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(54) Title: LIQUID CASTING COMPOSITIONS, PRODUCTION PROCESSES AND PHOTOCHROMIC OPTICAL ELEMENTS

(57) Abstract: The present invention provides a liquid casting composition for producing photochromic optical elements, the composition including: (i) an acrylate or methacrylate monomer; (ii) a photochromic compound; (iii) a terpinolene-based radical chain transfer agent; and (iv) a thermal free radical initiator. Also provided are methods for producing photochromic optical elements using the liquid casting composition and photochromic optical elements produced thereby. The photochromic optical elements include, for example, ophthalmic lenses.



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LIQUID CASTING COMPOSITIONS, PRODUCTION PROCESSES AND PHOTOCHROMIC OPTICAL ELEMENTS

Priority Claim

This application claims priority to Australian provisional patent application 2009901119 filed 16 March 2009, the contents of which are hereby incorporated by reference.

Field

The present invention relates to liquid casting compositions for producing photochromic optical elements, processes for producing optical elements containing the composition and photochromic optical elements produced using the compositions and/or processes.

Background

Photochromic optical elements, such as lenses, that darken and fade automatically in response to changing light are in widespread use. The optical elements contain photochromic compounds that change from a faded ground state to a darkened activated state upon exposure to sunlight. The transition from the ground state to the activated state is reversible so that the compounds revert to the transparent (or minimally coloured) ground state when removed from sunlight.

Photochromic optical elements are typically formed by including a photochromic compound within the substrate of the optical element, or on a surface of the optical element, and the optical elements are typically formed either by including the photochromic compound directly into the optical element substrate or by coating the optical element with a layer containing the photochromic compound. From a manufacturing point of view, it is preferable for the photochromic compound to be within the substrate of the optical element because it is cheaper to include the photochromic compound in the substrate during manufacture than it is to coat the optical element with a layer containing the photochromic compound.

The inclusion of a photochromic compound in the substrate of an optical element is usually achieved in one of two ways: (i) by imbuing the photochromic compound into a formed or semi-formed optical element (the so called 'imbibition' method); or (ii) by

including the photochromic compound into a casting composition which includes a polymerisable monomer, and then curing the composition to produce the optical element (the so called 'cast-in' method). In either case, it is widely recognised that inclusion of photochromic compounds into optical elements is difficult. For example, it is difficult to control the amount of photochromic compound introduced into an optical element using the imbibition method. In contrast, it is easier to control the amount of photochromic compound in the optical element using the cast-in method but the photochromic compound is often adversely affected by the polymerisation conditions with the result being degradation of the photochromic compound and poor photochromic performance of the resultant lens element.

A further consideration in the manufacture of photochromic optical elements is the performance of the photochromic compound when it is in the optical element substrate. Photochromic optical elements are generally expected to exhibit rapid conversion between faded and darkened states so that they react as rapidly as possible to any change in lighting conditions. It is known that the chemical and physical properties of the substrate matrix that surrounds the photochromic compound strongly influences the photochromic properties, such as the darkening and fading rate and the darkening depth.

Many existing processes for producing photochromic optical elements also result in sub-standard quality optical elements. For example, many processes result in optical elements that exhibit cracks and/or surface defects. This is often the case when the liquid casting composition is used to make optical elements of different thicknesses. For example, a composition which may be successfully used for a 2 mm plano lens may produce a 10 mm thick semifinished lens that is of sub-standard quality. It is therefore desirable to have a polymerisable composition that is robust enough to produce photochromic optical elements of different thicknesses without compromising the quality of the optical element.

From the foregoing description it is evident that many factors need to be considered in the manufacture of commercially viable photochromic optical elements. There is a need for improved and/or alternative liquid casting compositions and processes for forming polymeric photochromic optical elements.

Reference to any prior art in this specification is not, and should not be taken as, an acknowledgment or any form of suggestion that this prior art forms part of the common general knowledge in any country.

Summary

The present invention arises from our finding that by using a particular liquid casting composition it is possible to produce high quality, high performance photochromic optical elements.

The present invention provides a liquid casting composition for producing photochromic optical elements, the composition including:

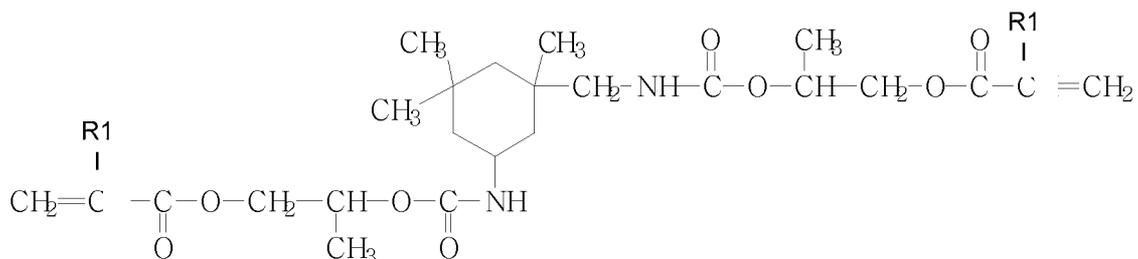
- (i) an acrylate or methacrylate monomer,
- (ii) a photochromic compound,
- (iii) a terpinolene-based radical chain transfer agent, and
- (iv) a thermal free radical initiator.

The term "liquid casting composition", as used herein, is intended to mean a polymerisable casting composition that may be added to a mould and subjected to a thermal polymerisation reaction to form a solid photochromic optical element.

A combination of an acrylate or methacrylate monomer and a terpinolene-based radical chain transfer agent is advantageous because the acrylate or methacrylate monomer is relatively easy to polymerise and, therefore, any effect of the photochromic compound on the polymerisation is minimised, and the terpinolene chain transfer agent modulates the polymerisation kinetics so that the optical element that is formed has adequate softness to allow the photochromic compound to switch rapidly between the darkened and faded states, but adequate hardness so that the optical element can be coated with coating layers. Furthermore, the terpinolene chain transfer agent controls the polymerisation reaction speed to achieve uniform and striation free optical elements.

In some embodiments, the acrylate or methacrylate monomer may include a urethane acrylate or methacrylate monomer. For example, the urethane acrylate or methacrylate monomer may include any one or more monomers selected from the group consisting of: NK Oligo U-4HA, NK Oligo U-6HA, and a monomer of the following structure:

- 4 -



wherein R1 represents a hydrogen atom or a methyl group.

The inclusion of a urethane acrylate or methacrylate monomer in the composition may improve one or more properties of an optical element formed from the composition. For example, the urethane acrylate or methacrylate monomer may improve the impact strength of the optical element that is formed.

The composition may also include a light stabiliser and/or a UV absorber and may also include an antioxidant.

Ideally, the composition will be suitable for forming photochromic optical elements with a Barcol hardness of between 60 and 85, between 60 and 80, or between 70 and 80. Barcol hardness is measured by using the Barcol hardness test, which measures the indentation hardness of materials through the depth of penetration of an indenter and comparison with a reference material. The test is known within the art and is governed by ASTM D 2583 (07 Standard Test Method for Indentation Hardness of Rigid Plastics by Means of a Barcol Impressor). In some embodiments, the Barcol hardness test may utilise a Model GYZJ 935 Barcol Testing Rig and Test Discs stamped 87/89 (GYZJ 69). These Test Discs have a Barcol reference hardness of 87/89 and are used as a reference for the test samples.

The composition may also be suitable for forming photochromic optical elements with a photochromic $T_{3/4}$ fade time less than 300 seconds or a photochromic $T_{3/4}$ fade time less than 250 seconds.

Further, the composition may be suitable for forming first and second crack-free and surface defect-free photochromic optical elements with centre thickness between 1.5 mm and 17 mm, wherein the centre thickness of the first optical element is at least 6 mm or at least 12 mm greater than that of the second optical element. In this regard, the same composition may be used to produce optical elements of different thickness ranges, which can simplify production. For example, in some embodiments, the same

composition may be used to produce any one or more of 2 mm plano lenses, -2.00 stock lenses, +4.00 stock lenses, 10 mm thick semi-finished lenses and 15 mm thick semifinished lenses.

The present invention also provides a process for the manufacture of a photochromic optical element, the process including:

- introducing the composition described above into a mould; and
- subjecting the composition to a thermal curing step to provide a solid photochromic optical element.

The present invention also includes an optical element formed from a composition as described herein or according to a process as described herein.

In some embodiments, the optical element is a lens, such as an ophthalmic lens.

General Description

As discussed, the present invention provides liquid casting compositions for producing photochromic optical elements, processes for producing optical elements containing the compositions and photochromic optical elements produced using the compositions and/or processes. The optical element may be any optical transparent article, such as lenses (including ophthalmic lenses and sunglass lenses), lens blanks, window panes, transparent display panels, and the like. In some embodiments, the optical element is a lens element. The term "lens element" as used herein refers to a finished or unfinished lens or lens blank manufactured from an optically transparent plastic material. In some embodiments, the lens element is an ophthalmic lens. Ophthalmic lenses can be used in sunglasses, fashion lenses, non-prescription (plano) lenses, prescription lenses (finished and semi-finished), sport masks, face shields and goggles. Whilst the present invention was developed in the context of manufacturing photochromic ophthalmic lenses, it will be evident that the processes and compositions described herein could also be used to form other transparent photochromic articles.

The optical element is formed from the composition, which includes (i) an acrylate or methacrylate monomer, (ii) a photochromic compound, (iii) a terpinolene-based radical chain transfer agent, and (iv) a thermal free radical initiator. The composition is

introduced into a mould and then thermally cured to provide a solid photochromic optical element.

The acrylate or methacrylate monomer may include one or more monomers selected from the group consisting of: alkylene diacrylates, such as decanediol diacrylate; poly(alkyleneoxide) diacrylates such as NK Ester A200 (polyethylene glycol 200 diacrylate), NK Ester A400 (polyethylene glycol 400 diacrylate), NK Ester A600 (polyethylene glycol 600 diacrylate), NK Ester APG-200 (tripropylene glycol diacrylate) and NK Ester APG-400 (polypropylene glycol 400 diacrylate); poly(alkyleneoxide) dimethacrylates such as NK Ester 14G, NK Ester 9G, 4g, and NK Ester 2G; and urethane acrylates or methacrylates such as NK Oligo U-4HA, NK Oligo U-6HA, NK Oligo U-2PPA. In some embodiments, the acrylate or methacrylate monomer is selected from the group consisting of: NK Ester 14G, NK Ester 9G, NK Ester 2G, and ethoxylated bisphenol-A dimethacrylate (having between 1 and 10 ethoxy groups per molecule). Optionally, the acrylate or methacrylate monomer also contains a urethane acrylate or methacrylate monomer such as NK Oligo U-4HA, NK Oligo U-6HA or NK Oligo U-2PPA.

It will be appreciated that the present invention contemplates compounds including a range of suitable acrylates and methacrylates. For example, the acrylate or methacrylate may include one or more selected from the group consisting of: Ethoxylated o-phenylphenol Acrylate, Methoxy Polyethylene Glycol 400 Acrylate(EO 9mol), Methoxy Polyethylene Glycol 550 Acrylate(EO 13mol), Phenoxy polyethyleneglycol Acrylate, Glycidyl Acrylate, Lauryl Methacrylate, Isostearyl Acrylate, 2-Hydroxy-1-Acryloxy-3-Methoxypropane, Polyethylene Glycol 200 Diacrylate, Polyethylene Glycol 400 Diacrylate, Polypropylene Glycol 600 Diacrylate, Propoxylated Ethoxylated Bis-A Diacrylate(PO12/EO6), 2.2 Bis[4-(AcryloxyPolyethoxy)Phenyl]Propane(EO3mol), 2.2Bis[4-(AcryloxyPolyethoxy)Phenyl]Propane(EO10mol), 2.2 Bis[4-(Acryloxy Polyethoxy)Phenyl]Propane(EO20mol), 2.2 Bis[4-(Acryloxy Diethoxy)Phenyl]Propane(EO4mol), 9,9-Bis[4-(2-acryloxy ethoxy)phenyl]fluorine, 2.2 Bis[4-(Acryloxy Polypropoxy)Phenyl]Propane(PO3mol), Tricyclodecane Dimethanol Diacrylate, 1,12-Dodecanediol Diacrylate, 1,10-Decanediol Diacrylate, 1,6-Hexanediol Diacrylate, 1,9-Nonanediol Diacrylate, Dipropylene Glycol Diacrylate, Tripropylene Glycol Diacrylate, Polypropylene Glycol #400 Diacrylate, Polypropylene Glycol #700 Diacrylate, Tris(2-acryloxyethyl) Isocyanurate, ϵ -Caprolactone ethoxylated isocyanuric acid triacrylate and Ethoxylated isocyanuric Acid

Triacrylate, Ethoxylated Glycerine Triacrylate(EO9mol), Ethoxylated Glycerine Triacrylate(EO20mol), Pentaerythritol Triacrylate, Trimethylol Propane Triacrylate, Ditrimehylol Propane Tetraacrylate, Ethoxylated Pentaerythritol tetraacrylate (EO35mol), Tetramethylol Methane Tetaacrylate, Di-pentaerythritol Polyacrylate, Di-pentaerythritol Polyacrylate, β -Methacryloyloxyethyl Hydrogen Phthalate, Methoxy Polyethylene Glycol 400 Methacrylate(EO 9mol), Methoxy Polyethylene Glycol 1000 Methacrylate(EO 23mol), Phenoxy Ethyl Methacrylate(EO 1mol), Glycidyl Methacrylate, Lauryl Methacrylate, Stearyl Methacrylate, Isostearyl Methacrylate, Ethylene Glycol Dimethacrylate, Diethylene Glycol Dimethacrylate, Triethyleneglycol Dimethacrylate, Polyethylene Glycol 200 Dimethacrylate, Polyethylene Glycol 400 Dimethacrylate, Polypropylene glycol 400 Dimethacrylate, Polypropylene Glycol 600 Dimethacrylate, Polyethylene Glycol 1000 Dimethacrylate, 2.2 Bis[4-(Methacryloxy Ethoxy)Phenyl]Propane(EO2.3mol), 2.2 Bis[4-(Methacryloxy Ethoxy)Phenyl]Propane (EO2.6mol), 2.2 Bis[4-(Methacryloxy Diethoxy)Phenyl]Propane(EO4mol), 2.2 Bis[4-(Methacryloxy Polyethoxy)Phenyl]Propane(EO10mol), 2.2 Bis[4-(Methacryloxy Polyethoxy)Phenyl]Propane (EO17mol), 2.2 Bis[4-(Methacryloxy Polyethoxy)Phenyl]Propane(EO30mol), Tricyclodecane Dimethanol Dimethacrylate, 1,12-Dodecanediol Dimethacrylate, 1.10-Decanediol Di Methacrylate, 1.6-Hexane Diol Dimethacrylate, 1.9-Nonanediol Di Methacrylate, Neopentyl Glycol Dimethacrylate, Ethoxylated Polypropyreneglycol Dimethacrylate(PO12/EO6), 2-Hydroxy 1,3-Dimethacryloxy Propane, Polypropylene Glycol #400 Dimethacrylate, Tris(2-methacryloxyethyl) Isocyanulate, ϵ -Caprolactone Ethoxylated Isocyanuric Acid Trimethacrylate, Ethoxylated Isocyanuric Acid Trimethacrylate, Ethoxylated Glycerine Trimethacrylate(EO9mol), Ethoxylated Glycerine Trimethacrylate(EO20mol), Pentaerythritol Trimethacrylate, Trimethylol Propane Trimethacrylate, Ditrimehylol Propane Tetramethacrylate, Ethoxylated Pentaerythritol Tetramethacrylate(EO35mol), Tetramethylol Methane Tetamethacrylate, Di-pentaerythritol Polymethacrylate, Di-pentaerythritol Polyacrylate, NK oligo U-4HA, NK oligo U-6HA, NK oligo U-200PA, NK oligo UA-122P, NK oligo UA-4200, NK oligo U-2PPA, Urethane Acrylate, Urethane Methacrylate, Epoxy Acrylate, Epoxy Methacrylate, Polybutadiene Dimethacrylate, Polybutadiene Diacrylate, and Polyester Acrylate.

Exemplary examples of the acrylate or methacrylate monomer include any one or more monomers selected from the group consisting of: NK Ester 14G, NK Ester 9G, NK Ester 2G, 1,12-Dodecanediol Dimethacrylate, 1.10-Decanediol Di Methacrylate, 1.9-Nonanediol Di Methacrylate (Light Ester 1.9 ND), Lauryl (meth)acrylate (NK Ester LA),

Isostearyl (meth)acrylate, and ethoxylated bisphenol-A dimethacrylate (having between 1 and 10 ethoxy groups per molecule).

The nature of the optical element substrate polymer can have an effect on the performance of photochromic compounds incorporated therein. For example, certain polymers may not have sufficient free volume or flexibility to allow the electrocyclic mechanism by which the photochromic compounds change colour to occur sufficiently. However, polymers that do have sufficient free volume or flexibility may not produce an optical element of sufficient hardness. As a result, it is difficult to produce photochromic optical elements having commercially acceptable photochromic speed and commercially acceptable hardness.

We have found that a combination of:

- (i) a relatively hard polymer formed from a polyalkylene oxide acrylate or methacrylate monomer;
- (ii) a relatively flexible polymer formed from a polyalkylene oxide acrylate or methacrylate monomer; and
- (iii) a urethane acrylate or methacrylate monomer

provides an optical element substrate having sufficient flexibility to allow for commercially acceptable photochromic speeds whilst also having commercially acceptable hardness and impact strength.

Examples of polyalkylene oxide acrylate or methacrylate monomers that form relatively hard polymers include those with low numbers of alkylene oxide units per monomer unit (e.g. monomers with 4 or less alkylene oxide units per monomer unit, such as diethyleneglycol dimethacrylate). Examples of polyalkylene oxide acrylate or methacrylate monomers that form relatively flexible polymers include those with high numbers of alkylene oxide units per monomer unit (e.g. 6 or more, 9 or more, or 14 or more alkylene oxide units per monomer unit, such as NK Ester 9G, NK Ester 14G and Light Ester 14 EG). It will be appreciated that some polyalkylene oxide acrylate or methacrylate monomers with alkylene oxide units within these specific ranges may not result in relatively hard or relatively flexible polymers and some monomers with alkylene oxide units outside of these specific ranges may result in relatively hard or relatively flexible polymers. However, a person skilled in the art would be able to determine whether a specific polyalkylene oxide acrylate or methacrylate monomer

forms a relatively hard or relatively flexible polymer by routine trial and error, especially by comparison with the examples provided.

Alternatively, we have found that a combination of:

- (i) a relatively flexible polymer formed from a polyalkylene oxide acrylate or methacrylate monomer; a polypropylene glycol acrylate or methacrylate monomer; a lauryl acrylate or methacrylate monomer; or a isostearyl acrylate or methacrylate monomer;
- (ii) a relatively hard polymer formed from a bisphenol A ethoxylated di acrylate or methacrylate; and
- (iii) a urethane acrylate or methacrylate monomer

provides an optical element substrate having sufficient flexibility to allow for commercially acceptable photochromic speeds whilst also having commercially acceptable hardness and impact strength.

Examples of polyalkylene oxide acrylate or methacrylate monomers that form relatively flexible polymers include those with high numbers of alkylene oxide units per monomer unit (e.g. 6 or more, 9 or more, or 14 or more alkylene oxide units per monomer unit, such as NK Ester 9G, NK Ester 14G and Light Ester 14 EG). It will be appreciated that some polyalkylene oxide acrylate or methacrylate monomers with alkylene oxide units within these specific ranges may not result in relatively flexible polymers and some monomers with alkylene oxide units outside of these specific ranges may result in relatively flexible polymers. However, a person skilled in the art would be able to determine whether a specific polyalkylene oxide acrylate or methacrylate monomer forms a relatively flexible polymer by routine trial and error, especially by comparison with the examples provided.

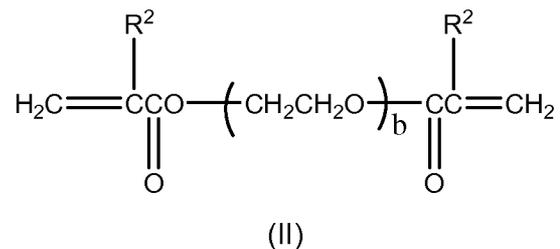
The ratios of monomers used may be varied to produce an optical element of a desired hardness.

As discussed above, the composition may be suitable for forming photochromic optical elements with a Barcol hardness of between 60 and 85, between 60 and 80, or between 70 and 80. Accordingly, the combination of monomers may be selected to provide an optical element with a Barcol hardness that falls within any of these ranges.

An example of a composition in accordance with the present invention may include:

- 10 -

- 30 to 60 parts by weight of a polymerisable composition polyethylene glycol di(meth)acrylate monomer of formula (II):

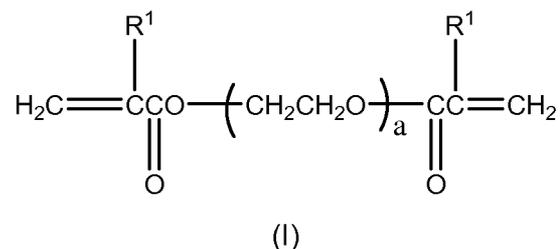


wherein R² is a hydrogen atom or a methyl group, and b is an integer selected from the group consisting of 5 to 20,

- 30 to 60 parts by weight of a bisphenol A ethoxylated di(meth)acrylate;
- 0 to 40 parts by weight of the polymerisable composition of a urethane (meth)acrylate monomer;
- 0.02 to 0.2 parts by weight of the polymerisable composition of the photochromic compound;
- 0.1 to 1.0 parts by weight of the polymerisable composition of the terpinolene-based radical chain transfer agent; and
- 0.1 to 1.5 parts by weight of the polymerisable composition of the thermal free radical initiator.

Alternatively, the composition may include:

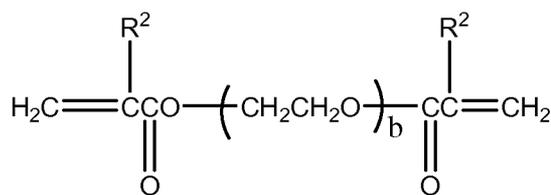
- 20 to 60 parts by weight of the polymerisable composition of a polyethylene glycol di(meth)acrylate monomer of formula (I):



wherein R¹ is a hydrogen atom or a methyl group, and a is an integer selected from the group consisting of 2 to 4;

- 15 to 60 parts by weight of the polymerisable composition of a polyethylene glycol di(meth)acrylate monomer of formula (II):

- 11 -



(II)

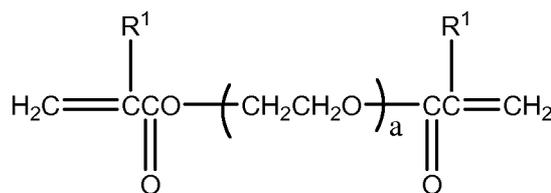
wherein R^2 is a hydrogen atom or a methyl group, and b is an integer selected from the group consisting of 5 to 20;

- 0 to 40 parts by weight of the polymerisable composition of a urethane (meth)acrylate monomer;
- 0.02 to 0.2 parts by weight of the polymerisable composition of the photochromic compound;
- 0.1 to 1.0 parts by weight of the polymerisable composition of the terpinolene-based radical chain transfer agent; and
- 0.1 to 1.5 parts by weight of the polymerisable composition of the thermal free radical initiator.

The composition may further include 5 to 20% parts by weight of the polymerisable composition of 1.9 nonanediol dimethacrylate or may further include 5 to 20% parts by weight of the polymerisable composition of n-Lauryl methacrylate.

As a further example, the composition may include:

- 30 to 50 parts by weight of the polymerisable composition of a polyethylene glycol di(meth)acrylate monomer of formula (I):

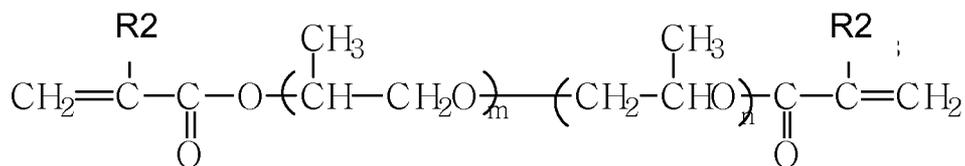


(I)

wherein R^1 is a hydrogen atom or a methyl group, and a is an integer selected from the group consisting of 2 to 4;

- 30 to 60 parts by weight of the polymerisable composition of a polypropylene glycol di(meth)acrylate monomer of formula (II):

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(II)

wherein R2 represents a hydrogen atom or a methyl group, and m and n are integers that together add up to 7;

- 0 to 40 parts by weight of the polymerisable composition of a urethane (meth)acrylate monomer;
- 0.02 to 0.2 parts by weight of the polymerisable composition of the photochromic compound;
- 0.1 to 1.0 parts by weight of the polymerisable composition of the terpinolene-based radical chain transfer agent; and
- 0.1 to 1.5 parts by weight of the polymerisable composition of the thermal free radical initiator.

Now that some exemplary examples of compounds have been described, attention will be directed to describing some of the components of the compounds in detail.

The photochromic compound may be chosen from a range of compounds that are known and have been used in applications in which a sunlight-induced reversible colour change or darkening is desired. Known chemical classes of photochromic compounds include naphthopyrans, anthraquinones, phthalocyanines, spiro-oxazines, chromenes, pyrans, fulgides, spiro-naphthoxazines, spiropyrans, triarylmethanes, stilbenes, azastilbenes, nitrones, spiropyrans, spiro-oxazines, quinones. Typically, the photochromic compounds have a visible lambda max of from 400 nm to 700 nm.

Examples of photochromic compounds may be selected from the group consisting of:

- 1,3-dihydrospiro[2H-anthra[2,3-d]imidazole-2,1'-cyclohexane]-5,10-dione;
- 1,3-dihydrospiro[2H-anthra[2,3-d]imidazole-2,1'-cyclohexane]-6,11-dione;
- 1,3-dihydro-4-(phenylthio)spiro[2H-anthra-1',2-diimidazole-2,1'-cyclohexane-6,11-dione];
- 1,3-dihydrospiro[2-H-anthra[1,2-d]imidazole-2,1'-cycloheptane]-6,11-dione-
- 1,3,3-trimethylspiroindole-2,3'-[3H]naphtho[2,1-b]-1,4-oxazine]
- 2-methyl-3,3'-spiro-bi-[3H-naphtho[2,1-bipyran](2-Me);
- 2-phenyl-3-methyl-7-methoxy-8'-nitrospiro[4H]-1-benzopyran-4,3'-[3H]-naphtho [2,1-

b]pyran;
 spiro[2H-1-benzopyran-2,9'-xanthene];
 8-methoxy-1',3'-dimethylspiro(2H-1-benzopyran-2,2'-(1'H)-quinoline;
 2,2'-spiro-bi-[2H-1-benzopyran];
 5'-amino-1',3',3'-trimethylspiro[2H-1-benzopyran-2,2'-indoline;
 ethyl-.beta.-methyl-.beta.-(3',3'-dimethyl-6-nitrospiro(2H-1-benzopyran-2,2'-indolin-1'-yl)-propenoate;
 (1,3-propanediyl)bis[3',3'-dimethyl-6-nitrospiro[2H-1-benzopyran-2,2'-indoline];
 3,3'-dimethyl-6-nitrospiro[2H-1-benzopyrao-2,2'-benzoxazoline];
 6'-methylthio-3,3'-dimethyl-8-methoxy-6-nitrospiro[2H-1-benzopyran-2,2'-benzothiozoline];
 (1,2-ethanediyl)bis[8-methoxy-3-methyl-6-nitrospiro[2H-1-benzopyran-2,2'-benzothiozoline];
 N-N'-bis(3,3'-dimethyl-6-nitrospiro[2H-1-benzopyran-2,2'(3'H)-benzothioazol-6'-yl)decanediamide];
 alpha.-(2,5-dimethyl-3-furyl)ethylidene(Z)-ethylidenesuccinicanhydride;
 .alpha.-(2,5-dimethyl-3-furyl)-.alpha.'.delta.-dimethylfulgide;
 2,5-diphenyl-4-(2'-chlorophenyl)imidazole;
 (2',4'-dinitrophenyl)methyl]-1H-benzimidazole;
 N-N-diethyl-2-phenyl-2H-phenanthro[9,10-d]imidazol-2-amine;
 2-nitro-3-aminofluoren 2-amino-4-(2'-furanyl)-6H-1,3-thiazine-6-thione.

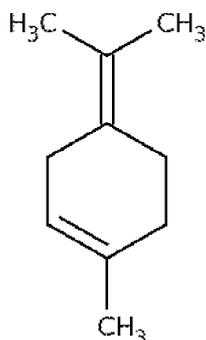
Commercial photochromic compounds may also be used. Examples of commercial photochromic compounds may include any one or more selected from the group consisting of: CNN11, CNN12, CNN13, CNN14, CNN15, CNN16, CNN17 (from Tokuyama), Reversacol Midnight Gray, Reversacol Pacific Blue, Reversacol Sunflower, and Reversacol Corn Yellow (from James Robinson). Other known commercial photochromic compounds may also be used.

Optionally, a mixture of two or more of the photochromic compounds may be used. By using appropriate mixtures of photochromic compounds, it is possible to obtain specific activated colours.

The terpinolene-based radical chain transfer agent functions as a polymerisation moderator and, as such, it helps to minimise the formation of distortions, such as striations, in the optical elements that are produced. The term "terpinolene-based

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radical chain transfer agent” as used herein is intended to include within its scope terpinolene itself and structurally related analogues of terpinolene, such as 1-isopropyl-4-methyl-1,4-cyclohexadiene and 1-isopropyl-4-methyl-1,3-cyclohexadiene. Terpinolene, which has a CAS Registry ID of 586-62-9, has the following structure:



The terpinolene-based radical chain transfer agent may also be a mixture of compounds. The terpinolene-based radical chain transfer agent may be added to the polymerisable composition in an amount from 0.01 percent to 2 percent by weight based on the total weight of the polymerisable organic composition. The skilled person will appreciate that the actual amount of terpinolene-based radical chain transfer agent used can be determined taking into account a number of factors, including the specific acrylate or methacrylate monomers used, the thermal free radical initiator used, the photochromic compound, as well as the amounts of each of the components in the polymerisable composition.

We have observed that polymerisation of compositions containing terpinolene results in the formation of optical elements that are uniform when tested under standard test conditions (i.e. polariscope and shadowscope) and also show good photochromic speeds. In contrast, polymerisation of compositions that do not contain terpinolene results in the formation of optical elements that are not uniform under the same test conditions and/or have slower photochromic speeds.

Thermal free radical initiators are heat activated catalysts that initiate the polymerisation reaction. A number of thermal free radical initiators are known in the art and can be used. Examples include: azo free radical initiators such as AIBN (azodiisobutyronitrile), 2,2'-Azobis(4-methoxy-2,4-dimethyl valeronitrile), 2,2'-Azobis(2,4-dimethyl valeronitrile), Dimethyl 2,2'-azobis(2-methylpropionate), 2,2'-Azobis(2-methylbutyronitrile), 1,1'-Azobis(cyclohexane-1-carbonitrile), 2,2'-Azobis[N-(2-propenyl)-2-methylpropionamide]; dialkyl peroxide free radical initiators such as 1,1-di-(butylperoxy-3,3,5-trimethyl cyclohexane); alkyl perester free radical initiators such as

TBPEH (t-butyl per-2-ethylhexanoate); diacyl peroxide free radical initiators such as benzoyl peroxide; peroxy dicarbonate radical initiators such as ethyl hexyl percarbonate; ketone peroxide initiators such as methyl ethyl ketone peroxide, bis(t-butyl peroxide) diisopropylbenzene, t-butylperbenzoate, t-butyl peroxy neodecanoate, and combinations thereof. Further examples of organic peroxide free radical initiators include: Dilauroyl peroxide, 2,2-Di(4,4-di(tert-butylperoxy)cyclohexyl)propane, Di(tert-butylperoxyisopropyl)benzene, Di(4-tert-butylcyclohexyl) peroxydicarbonate, Dicycetyl peroxydicarbonate, Dimyristyl peroxydicarbonate, 2,3-Dimethyl-2,3-diphenylbutane, Dicumyl peroxide, Dibenzoyl peroxide, Diisopropyl peroxydicarbonate, tert-Butyl monoperoxymaleate, 2,5-Dimethyl-2,5-di(tert-butylperoxy)hexane, tert-Butylperoxy 2-ethylhexyl carbonate, tert-Amyl peroxy-2-ethylhexanoate, tert-Amyl peroxy pivalate, tert-Amylperoxy 2-ethylhexyl carbonate, 2,5-Dimethyl-2,5-di(2-ethylhexanoylperoxy)hexane, 2,5-Dimethyl-2,5-di(tert-butylperoxy)hexyne-3, Di(3-methoxybutyl) peroxydicarbonate, Diisobutyl peroxide, tert-Butyl peroxy-2-ethylhexanoate (Trigonox 21S), 1,1-Di(tert-butylperoxy)cyclohexane, tert-Butyl peroxyneodecanoate, tert-Butyl peroxy pivalate, tert-Butyl peroxyneohexanoate, tert-Butyl peroxydiethylacetate, 1,1-Di(tert-butylperoxy)-3,3,5-trimethylcyclohexane, 3,6,9-Triethyl-3,6,9-trimethyl-1,4,7-triperoxonane, Di(3,5,5-trimethylhexanoyl) peroxide, tert-Butyl peroxy-3,5,5-trimethylhexanoate, 1,1,3,3-Tetramethylbutyl peroxy-2-ethylhexanoate, 1,1,3,3-Tetramethylbutyl peroxyneodecanoate, tert-Butyl peroxy-3,5,5-trimethylhexanoate, Cumyl peroxyneodecanoate, Di-tert-butyl peroxide, tert-Butylperoxy isopropyl carbonate, tert-Butyl peroxybenzoate, Di(2-ethylhexyl) peroxydicarbonate, tert-Butyl peroxyacetate, Isopropylcumyl hydroperoxide, and tert-Butyl cumyl peroxide.

The composition and/or the thermally cured optical element may contain additional conventional adjuvants that impart desired properties or characteristics to the optical element, or which enhance the performance of the optical element. Such adjuvants include, but are not limited to, light stabilisers, UV absorbers, heat stabilisers, tints, dyes, free radical scavengers, plasticisers, flow additives, antioxidants, and other processing aids. UV absorbers may include, for example, members or derivatives of a benzophenone class, a benzotriazole class, a cyanoacrylate class or a triazine class. Commercially available UV absorbers may include, for example Chimassorb 81 or Tinuvin 81/213/234/326/328/329/360/571/1577/P (from Ciba).

The composition may further include a light stabiliser. Hindered amine light stabilisers (HALS) are available commercially. HALS are derivatives of 2,2,6,6-tetramethyl

piperidine and are extremely efficient stabilizers against light-induced degradation of most polymers and photochromic dyes. An exemplary example of a suitable light stabiliser is Tinuvin 765, which is available from Ciba. Other examples of HALS include Chimassorb 2020/944, Tinuvin 111/123/494/622/770/783/791/B75/NOR 371/XT 833/XT 850.

The polymerisable composition may further include one or more antioxidants. In some embodiments, the one or more antioxidants may improve the quality and/or durability of the optical element formed from the polymerisable composition. The one or more antioxidants may include a primary antioxidant (e.g. a butylated hydroxytoluene (BHT) or a hindered phenol) and/or a secondary antioxidant (e.g. a phosphite or thioester). Commercially available antioxidants include, for example, Irganox 1010, Irganox 1076, Irganox 245, Irganox 1035, Irganox B225, Irganox B900, Irgafos 126, Irgafos P-EPQ, Irgafos 168, and Irgafos 38. The antioxidant may be used in the polymerisable composition in a concentration of 0.01 to 0.5 parts by weight of the polymerisable composition. Preferably, the antioxidant will be used in the polymerisable composition in a concentration of approximately 0.1 parts by weight of the polymerisable composition.

The composition may further include a small quantity of a permanent dye (i.e. a non photochromic dye such as Solvaperm red BB). The small quantity of permanent dye may be used to minimise or mask undesired residual colour in the optical element formed from the polymerisable composition. For example, 0.01 to 0.1 parts of a 0.025% solution of a permanent dye concentrate may be used in the polymerisable composition.

The present invention also includes an optical element formed from the compositions described herein and/or produced according to any of the methods described herein. In some embodiments, the optical element is a lens (e.g. an ophthalmic lens).

The optical element may be formed by introducing the composition into an assembled mould. Methods for moulding plastic optical elements are known in the art. Prior to introducing the polymerisable composition into the mould it may be degassed. The polymerisable composition may also be filtered prior to introduction into the mould in order to remove any particulate matter that may affect the quality of the thermally cured optical element.

After the polymerisable composition has been introduced into the mould the composition is cured to form a solid optical element. In some embodiments, carefully controlling the polymerisation step allows for the production of high quality, high performance photochromic optical elements. The curing is carried out under thermal conditions, i.e. by heating the assembled mould containing the polymerisable composition and may include using a thermal gradient. For example, the thermal gradient may begin with a time period at about 50°C, after which the temperature is increased incrementally over a period of about 12 hours to a final temperature of about 120°C, and then decreased to about 70°C or about 80°C.

Accordingly, the thermal curing step may include raising the temperature of the composition from around 50°C to between 80°C and 120°C over a period of 7 to 14 hours. Optionally, the thermal curing step may include as a final step, maintaining the temperature of the composition at between about 70°C and 80°C for 0.5 to 2 hours.

An exemplary the thermal curing step may include:

- heating the composition at 52°C for 3 hours;
- heating the composition from 52°C to 53.5°C over 1.5 hours;
- heating the composition from 53.5°C to 62.5°C over 4.5 hours;
- heating the composition from 62.5°C to 80°C over 4 hours; and
- maintaining the temperature at 80°C for 8 hours.

It will be appreciated that minor variations to the above thermal curing step may also be contemplated and are within the scope of the present invention.

After cooling, the cured optical element may be removed from the mould and subjected to further processes, if desired. For example, other coatings known in the art may be coated onto the optical element. For example, one or more coatings that provide abrasion and scratch resistance can be coated onto the optical element. Suitable abrasion resistant coating materials are known in the art and include polyfunctional acrylic hard coatings, urethane-based hard coatings, alkyl-based coatings, siloxane based hard coatings or other organic or inorganic/organic hard coatings.

The term "photochromic speed" as used herein in relation to the fading of a photochromic optical element, refers to the time taken for the optical density of the

optical element to fade to a predetermined fraction of an activated value. Accordingly, the term “ $T_{3/4}$ ”, as used herein in relation to fading of a photochromic optical element, refers to the time taken for the optical density of the lens to decrease by 75% from an activated state.

In some embodiments, optical elements produced according to the present invention may have a photochromic $T_{3/4}$ fade time of less than 300 seconds or a photochromic $T_{3/4}$ fade time of less than 250 seconds.

Description of Embodiments

Examples of materials and methods for use with the compositions and methods of the present invention will now be provided. In providing these examples, it is to be understood that the specific nature of the following description is not to limit the generality of the above description.

Each of the examples provided herein were used to produce optical elements from each of the following mould assemblies:

Mould assemblies
Moulds for 15 mm thick semifinished lens
Moulds for 10mm thick semifinished lens
Moulds for +4.00 stock lens
Moulds for -2.00 stock lens
Moulds for 2 mm plano

Example 1

A polymerisable composition containing the following components was prepared as described below:

Component Type	Component Name (commercial source)	Parts
Photochromic dye	Reversacol midnight gray (James Robinson)	0.13
Monomer	NK Ester 9G (Shin Nakamura Chemical)	45
Monomer	NS110 (Akzo Chemie)	50
Monomer	*NK Oligo U-4HA (Shin Nakamura Chemical)	5
Cure modifier	Terpinolene	0.4
Light stabiliser	**Tinuvin 765 (Ciba)	0.05
Thermal initiator	***Trigonox 21S (Akzo)	1.0

*NK Oligo U-4HA is an urethane monomer having 4 terminal acrylic or methacrylic groups. **Tinuvin 765 is a hindered Amine light stabilizer. ***Trigonox 21S is a thermal free radical initiator.

The photochromic dye was dissolved for 8 hours in the monomers before the thermal initiator was added. After degassing and filtration, the mould assemblies were filled with the monomer mix and cured in an oven with the following heating profile:

Curing step	Curing program
Step 1	Start at 52 degree C
Step 2	Hold at 52 degree C for 2 hours
Step 3	Heat up to 56 degree C in 4 hours
Step 4	Heat up to 60 degree C in 2 hours
Step 5	Heat up to 65 degree C in 2 hours
Step 6	Heat up to 80 degree C in 3.5 hours
Step 7	Heat up to 120 degree C in 1 hours
Step 8	Hold at 120 degree C for 2 hours
Step 9	Cool down to 70 degree C in 1 hour

The cured lenses from each mould assembly were uniform under polariscope and shadowscope. There were no optical distortions in the material and the photochromic speed was good. The lenses gave a bluish gray colour when activated in the sun light.

Example 2

A polymerisable composition containing the following components was prepared as described below:

Component Type	Component Name (commercial source)	Parts
Photochromic dye	Reversacol Pacific blue (James Robinson)	0.08
Photochromic dye	Reversacol Sunflower (James Robinson)	0.017
Monomer	NK Ester 2G (Shin Nakamura Chemical)	45
Monomer	Light Ester 14 EG (Kyoisha Chemical)	35
Monomer	NK Oligo U-2PPA (Shin Nakamura Chemical)	20
Cure modifier	Terpinolene	0.5
Light stabilizer	Tinuvin 765	0.05
Thermal initiator	Trigonox 21S	0.4

Of these components, Reversacol Pacific blue is a blue photochromic dye Reversacol Sunflower is a yellow photochromic dye, NK Ester 2G is a Diethylene glycol dimethacrylate, Light Ester 14 EG is a polyethylene glycol dimethacrylate And NK Oligo U-2PPA is a urethane diacrylate.

The photochromic dyes were dissolved in NK Ester 2G first for three hours, then the remaining components were added in and stirred. After degassing and filtration, the monomer mix was filled in the mould assemblies and cured in the oven with the following heating profile.

Curing step	Curing program
Step 1	Start at 52 degree C
Step 2	Hold at 52 degree C for 3 hours
Step 3	Heat up to 53.5 degree C in 1.5 hours
Step 4	Heat up to 62.5 degree C in 4.5 hours
Step 5	Heat up to 80 degree C in 4 hours
Step 6	Heat up to 90 degree C in 3.5 hours
Step 7	Hold at 90 degree C for 2 hours
Step 8	Cool down to 80 degree C in 1 hour
Step 9	Hold at 80 degree C for 1.5 hours

The cured lenses from each mould assembly were uniform under polariscope and shadowscope. There were no optical distortions in the material and the photochromic speed was good. The lenses gave a gray colour when activated in the sun light.

Example 3

A polymerisable composition containing the following components was prepared as described below:

Component Type	Component Name (commercial source)	Parts
Photochromic dye	Reversacol midnight gray (James Robinson)	0.13
Photochromic dye	Reversacol corn yellow (James Robinson)	0.04
Monomer	NK Ester 2G	40
Monomer	NK Ester 14G	40
Monomer	NK Oligo U-2PPA	20
Cure modifier	Terpinolene	0.5
Light stabilizer	Tinuvin 765	0.05
Thermal initiator	Trigonox 21S	1.0

Reversacol midnight gray is a blue photochromic dye and Reversacol corn yellow is a yellow photochromic dye.

The photochromic dyes were dissolved in NK Ester 2G first for three hours, then the remaining components were added in and stirred. After degassing and filtration, the monomer mix was filled in the mould assemblies and cured in the oven with the following heating profile:

Curing step	Curing program
Step 1	Start at 52 degree C
Step 2	Hold at 52 degree C for 3 hours
Step 3	Heat up to 53.5 degree C in 1.5 hours
Step 4	Heat up to 62.5 degree C in 4.5 hours
Step 5	Heat up to 80 degree C in 3.5 hours
Step 6	Hold at 80 degree C for 1 hours
Step 7	Heat up to 120 degree C in 5 hours
Step 8	Hold at 120 degree C for 2 hours
Step 9	Cool down to 80 degree C in 1 hour

The cured lenses from each mould assembly were uniform under polariscope and shadowscope. There were no optical distortions in the material and the photochromic speed was good. The lenses gave a gray colour when activated in the sun light.

Example 4

A polymerisable composition containing the following components was prepared as described below:

Component Type	Component Name	Parts
Photochromic dye	Reversacol Pacific blue	0.08
Photochromic dye	Reversacol sunflower	0.017
Monomer	NK Ester 2G	36
Monomer	Light Ester 14 EG	31
Monomer	NK Oligo U-2PPA	33
Cure modifier	Terpinolene	0.5
Light stabiliser	Tinuvin 765	0.05
Thermal initiator	Trigonox 21S	0.4

The photochromic dyes were dissolved in NK Ester 2G first for three hours, then the remaining components were added in and stirred. After degassing and filtration, the monomer mix was filled in the mould assemblies and cured in an oven with the following heating profile:

Curing step	Curing program
Step 1	Start at 52 degree C
Step 2	Hold at 52 degree C for 3 hours
Step 3	Heat up to 53.5 degree C in 1.5 hours
Step 4	Heat up to 62.5 degree C in 4.5 hours
Step 5	Heat up to 80 degree C in 4 hours
Step 6	Heat up to 90 degree C in 3.5 hours
Step 7	Hold at 90 degree C for 2 hours
Step 8	Cool down to 80 degree C in 1 hour
Step 9	Hold at 80 degree C for 1.5 hours

The cured lenses from each mould assembly were uniform under polariscope and shadowscope. There were no optical distortions in the material and the photochromic speed was good. The lenses gave a gray colour when activated in the sun light.

Example 5

A polymerisable composition containing the following components was prepared as described below:

Component Type	Component Name (commercial source)	Parts
Photochromic dye	Reversacol Pacific blue (James Robinson)	0.08
Photochromic dye	Reversacol sunflower (James Robinson)	0.017
Monomer	NK Ester 2G (Shin Nakamura Chemical)	50
Monomer	Light Ester 14 EG (Kyoisha Chemical)	30
Monomer	NK Oligo U-2PPA (Shin Nakamura Chemical)	20
Cure modifier	Terpinolene	0.5
Light stabiliser	Tinuvin 765	0.05
Thermal initiator	Trigonox 21S	0.4

Of these components: Reversacol Pacific blue is a blue photochromic dye; Reversacol Sunflower is a yellow photochromic dye; NK Ester 2G is a Diethylene glycol dimethacrylate; Light Ester 14 EG is a polyethylene glycol dimethacrylate; and NK Oligo U2 PPA is a urethane diacrylate.

The photochromic dyes were dissolved in NK Ester 2G first for three hours, then the remaining components were added in and stirred. After degassing and filtration, the monomer mix was filled in the moulds assemblies and cured in the oven with the following heating profile:

Curing step	Curing program
Step 1	Start at 52 degree C
Step 2	Hold at 52 degree C for 3 hours
Step 3	Heat up to 53.5 degree C in 1.5 hours
Step 4	Heat up to 62.5 degree C in 4.5 hours
Step 5	Heat up to 80 degree C in 4 hours
Step 6	Heat up to 90 degree C in 3.5 hours
Step 7	Hold at 90 degree C for 2 hours
Step 8	Cool down to 80 degree C in 1 hour
Step 9	Hold at 80 degree C for 1.5 hours

The cured lenses from each mould assembly were uniform under polariscope and shadowscope. There were no optical distortions in the material and the photochromic speed was good. The lenses gave a gray colour when activated in the sun light.

Example 6

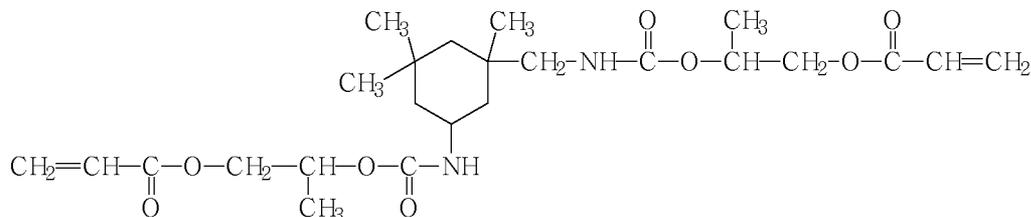
A polymerisable composition containing the following components was prepared as described below:

Component Type	Component Name (commercial source)	Parts
Photochromic dye	CNN16 (Tokuyama)	0.013
Photochromic dye	CNN12 (Tokuyama)	0.0075
Photochromic dye	CNN13 (Tokuyama)	0.04
Photochromic dye	CNN15 (Tokuyama)	0.0075
Photochromic dye	CNN14 (Tokuyama)	0.0075
Cure modifier	Terpinolene	0.5
Light Stabiliser	*Tinuvin 765 (CIBA)	0.05
Permanent dye	red dye concentrate including 0.025% Solvaperm red BB (Clariant) in NK ester 2G	0.04
monomer	NK ester 2G	50
monomer	NK ester 14G	30
monomer	**NK Oligo U-2PPA (Shin Nakamura Chemical)	20
Thermal initiator	Trigonox 21S	0.31

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*Tinuvin 765 is a hindered amine light stabiliser (HALS) and is a mixture of Bis(1,2,2,6,6-pentamethyl-4-piperidyl)sebacate and methyl 1,2,2,6,6-pentamethyl-4-piperidyl sebecate.

**NK Oligo U-2PPA is a monomer, which has following chemical structure:



The monomer mix was prepared based on the above formulation. After stirring, degassing and filtration the monomer mix was filled into the mould/gasket assemblies.

The assemblies were placed in an oven to be cured using the following curing profile:

Curing step	Curing program
Step 1	Start at 52 °C
Step 2	Hold at 52 °C for 3 hours
Step 3	Heat up from 52 to 53.5 °C in 1.5 hours
Step 4	Heat up from 53.5 to 62.5 °C in 4.5 hours
Step 5	Heat up from 62.5 to 80 °C in 4 hours
Step 6	Hold at 80 °C for 8 hours

This formulation and curing profile were suitable for producing thick semifinished lens blanks and thin stock lenses. The produced lenses from each mould assembly had good quality and the casting yield was high. The lenses are gray photochromic lenses and the basic material properties of such produced lenses are as follows:

Material properties	Values
Refractive Index (d line)	1.5046
Abbe number	54
Density	1.21 g/cm ³
Shrinkage	10%
Monomer Viscosity	72 cps
Rimless tensile strength (2 mm planos)	20.9 kg

Impact strength (2.2 mm, -2.00 lenses, AR coated, no primer)	Passed FDA impact test
Static load test (1.8 mm CT)	Pass
Heat deflection temperature	58.8 °C
Tg	112 °C
Barcol Hardness	76.2
Photochromic properties	
Initial T%	90.2%
Fully activated T%	10.6%
Photochromic speed, T _{3/4} fade (seconds)	200 seconds

In this example, and the following examples, initial T% and fully activated T% relate to optical transmission test results of the optical elements maintained in dark or light conditions. The tests were performed as specified in EN1836:1997 and ISO 8980-3, with a conditioning beam (as specified in the standard) being set to 50klux. All lenses were tested at 23 °C.

Example 7

A polymerisable composition containing the following components was prepared as described below:

Component Type	Component Name	Parts
Photochromic dye	CNN16	0.013
Photochromic dye	CNN12	0.0075
Photochromic dye	CNN13	0.04
Photochromic dye	CNN15	0.0075
Photochromic dye	CNN14	0.0075
Cure modifier	Terpinolene	0.5
Light Stabiliser	Tinuvin 765	0.05
Permanent dye	red dye concentrate(0.025% Solvaperm red BB in NK ester 9G)	0.03
monomer	NK ester 2G	30
monomer	*Light Ester 1.9ND (Kyoieisha Chemical)	10
monomer	NK ester 9G	30
monomer	NK Oligo U-2PPA	30
Thermal initiator	Trigonox 21S	0.31

*Light Ester 1.9 ND is a monomer with the chemical name of 1.9 Nonanediol dimethacrylate

The monomer mix was prepared based on the above formulation. After stirring, degassing and filtration the monomer mix was filled into the mould/gasket assemblies. The assemblies were placed in an oven to be cured using the following curing profile:

Curing step	Curing program
Step 1	Start at 52 oC
Step 2	Hold at 52 oC for 3 hours
Step 3	Heat up from 52 to 53.5 oC in 1.5 hours
Step 4	Heat up from 53.5 to 62.5 oC in 4.5 hours
Step 5	Heat up from 62.5 to 80 oC in 4 hours
Step 6	Hold at 80 oC for 8 hours

This formulation and curing profile were suitable for producing thick semifinished lens blanks and stock lenses. The produced lenses from each mould assembly had good quality and the casting yield was high. The lenses are gray photochromic lenses and the basic material properties of such produced lenses are as follows:

Material properties	Value
Refractive Index (d line)	1.5020
Abbe number	54
Density	1.20 g/cm ³
Shrinkage	10.5%
Monomer Viscosity	47 cps
Rimless tensile strength (2 mm planos)	22 kg
Impact strength (2.2 mm, -2.00, AR no primer)	Passed FDA impact test
Static load test (1.8 mm CT)	pass
Heat deflection temperature	57.3 °C
Tg	94 °C
Barcol Hardness	76.5
Photochromic properties	
Initial T%	89.6%
Fully activated T%	8.4%
Photochromic speed, T _{3/4} fade (seconds)	211 seconds

Example 8

A polymerisable composition containing the following components was prepared as described below:

Component Type	Component Name	Parts
Photochromic dye	CNN16	0.013
Photochromic dye	CNN12	0.005
Photochromic dye	CNN13	0.04
Photochromic dye	CNN15	0.0075
Photochromic dye	CNN14	0.01
Cure modifier	Terpinolene	0.5
Light Stabiliser	Tinuvin 765	0.05
Permanent dye	red dye concentrate(0.025% Solvaperm red BB in NK ester 9G)	0.03
monomer	NK ester 2G	35
monomer	NK ester 9G	20
monomer	*NK ester LA (Shin-Nakamura Chemical)	10
monomer	NK Oligo U-2PPA	35
Thermal initiator	Trigonox 21S	0.4

*NK ester LA is a monomer with the chemical name of n-Lauryl methacrylate.

In this formulation, a mono functional methacrylate n-Lauryl methacrylate was used. The monomer mix was prepared using above formulation. After stirring, degassing and filtration, the monomer mix was filled into the mould assemblies. The mould assemblies were placed in an oven and cured using the following curing profile:

Curing step	Curing program
Step 1	Start at 52 °C
Step 2	Hold at 52 °C for 3 hours
Step 3	Heat up from 52 to 53.5 °C in 1.5 hours
Step 4	Heat up from 53.5 to 62.5 °C in 4.5 hours
Step 5	Heat up from 62.5 to 80 °C in 4 hours
Step 6	Hold at 80 °C for 8 hours

The lenses generated in this method from each mould assembly have good and balanced lens properties. Both thick semi-finished and thin stock lenses can be cast using this method and formulation without problems. The lenses are gray photochromic lenses and the basic material properties of such achieved lenses are listed in following table:

Material properties	Value
Refractive Index (d line)	1.501

The monomer mix was prepared using above formulation. After stirring, degassing and filtration, the monomer mix was filled into the moulds assemblies. The moulds assemblies were placed in an oven and cured using the following curing profile:

Curing step	Curing program
Step 1	Start at 52 °C
Step 2	Hold at 52 °C for 3 hours
Step 3	Heat up from 52 to 53.5 °C in 1.5 hours
Step 4	Heat up from 53.5 to 62.5 °C in 4.5 hours
Step 5	Heat up from 62.5 to 80 °C in 4 hours
Step 6	Hold at 80 °C for 8 hours

The lenses generated in this method from each mould assembly have good and balanced lens properties. Both thick semi-finished and thin stock lenses can be cast using this method and formulation without problems. The lenses are gray photochromic lenses and the basic material properties of such achieved lenses are listed in following table:

Material properties	Value
Refractive Index (d line)	1.4971
Abbe number	56
Density	1.168 g/cm ³
Rimless tensile strength (2 mm planos)	21 kg
Impact strength (2.2 mm, -2.00, AR coated, no primer)	Pass FDA impact testing
Static load test (1.8 mm CT)	pass
Heat deflection temperature	62.1 °C
Tg	100.5 °C
Barcol Hardness	77.6
Photochromic properties	
Initial T%	89.3%
Fully activated T%	8.0%
Photochromic speed, T _{3/4} fade (seconds)	209 seconds

Example 10

A polymerisable composition identical to Example 7 was produced, except that it contained 0.005% extra UV absorber Tinuvin 329 (from CIBA). Lenses were formed following the methods set out in Example 7.

The chemical name for the UV absorber used in this example is 2-(2H-benzotriazol-2-yl)-4-(1,1,3,3-tetramethylbutyl)phenol.

The resulting lenses from each mould assembly were gray photochromic lenses and the quality was good. The photochromic performance of the lenses is as follows:

Initial T%	90.1%
Fully activated T%	12.4%
Photochromic speed, $T_{3/4}$ fade (seconds)	164 seconds

The resulting lenses had a Barcol hardness of 78.7.

Example 11

A polymerisable composition containing the following components was prepared as described below:

Component Type	Component Name	Parts
Photochromic dye	CNN16	0.025
Photochromic dye	CNN17 (Tokuyama)	0.005
Photochromic dye	CNN11 (Tokuyama)	0.0125
Photochromic dye	CNN12	0.00625
Photochromic dye	CNN13	0.0375
Cure modifier	Terpinolene	0.5
Light Stabiliser	Tinuvin 765	0.05
monomer	NK ester 2G	30
monomer	Light Ester 1.9ND	10
monomer	NK ester 9G	30
monomer	NK Oligo U-2PPA	30
Thermal initiator	Trigonox 21S	0.31

In this formulation, the brown dye package from Tokuyama was used.

The monomer mix was prepared based on the above formulation. After stirring, degassing and filtration the monomer mix was filled into the mould/gasket assemblies. The assemblies were placed in the oven to be cured using the following curing profile:

Curing step	Curing program
Step 1	Start at 52 °C
Step 2	Hold at 52 °C for 3 hours
Step 3	Heat up from 52 to 53.5 °C in 1.5 hours
Step 4	Heat up from 53.5 to 62.5 °C in 4.5 hours
Step 5	Heat up from 62.5 to 80 °C in 4 hours
Step 6	Hold at 80 °C for 8 hours

This formulation and curing profile were suitable for producing thick semi-finished lens blanks and stock lenses. The lenses produced from each mould assembly were brown photochromic lenses, which were of good quality and the casting yield was high. The photochromic properties of the lenses are as follows:

Initial T%	90.7%
Fully activated T%	13.8%
Photochromic speed, T3/4 fade (seconds)	156 seconds

The resulting lenses had a Barcol hardness of 76.2.

Comparative Example 1

A polymerisable composition containing the following components was prepared as described below:

Component Type	Component Name	Parts
Photochromic dye	Reversacol midnight gray	0.13
Monomer	NK Ester 9G	45
Monomer	NS110	50
Monomer	NK Oligo U4HA	5
Light stabilizer	Tinuvin 765	0.05
Thermal initiator	Trigonox 21S	1.0

The photochromic dye was dissolved for 8 hours in the monomers before the thermal initiator was added. After degassing and filtration, the mould assemblies were filled with the monomer mix and cured in an oven with the following heating profile used:

Curing step	Curing program
Step 1	Start at 52 degree C
Step 2	Hold at 52 degree C for 2 hours
Step 3	Heat up to 56 degree C in 4 hours
Step 4	Heat up to 60 degree C in 2 hours

Step 5	Heat up to 65 degree C in 2 hours
Step 6	Heat up to 80 degree C in 3.5 hours
Step 7	Heat up to 120 degree C in 1 hours
Step 8	Hold at 120 degree C for 2 hours
Step 9	Cool down to 70 degree C in 1 hour

This comparative example did not use terpinolene in the formulation. The lenses obtained were not uniform under polariscope and shadowscope and there were a lot of strains in the lenses. They were not considered to be good lenses.

Comparative Example 2

A polymerisable composition containing the following components was prepared as described below:

Component Type	Component Name	Parts
Photochromic dye	Reversacol Pacific blue	0.08
Photochromic dye	Reversacol sunflower	0.017
Monomer	NK Ester 2G	45
Monomer	Light Ester 14 EG	35
Monomer	NK Oligo U-2PPA	20
Light stabilizer	Tinuvin 765	0.05
Thermal initiator	Trigonox 21S	0.4

The photochromic dyes were dissolved in NK Ester 2G first for three hours, then the remaining components were added in and stirred. After degassing and filtration, the monomer mix was filled in the mould assemblies and cured in the oven with the following heating profile:

Curing step	Curing program
Step 1	Start at 52 degree C
Step 2	Hold at 52 degree C for 3 hours
Step 3	Heat up to 53.5 degree C in 1.5 hours
Step 4	Heat up to 62.5 degree C in 4.5 hours
Step 5	Hold at 90 degree C for 2 hours
Step 6	Cool down to 80 degree C in 1 hour
Step 7	Hold at 80 degree C for 1.5 hours

This comparative example did not use terpinolene in the formulation. The lenses obtained were not uniform under polariscope and shadowscope and there were a lot of strains in the lenses. They were not considered to be good lenses.

Comparative Example 3

A polymerisable composition containing the following components was prepared as described below:

Component Type	Component Name (commercial source)	Parts
Photochromic dye	Reversacol midnight gray (James Robinson)	0.13
Photochromic dye	Reversacol corn yellow (James Robinson)	0.04
Monomer	NK Ester 2G	40
Monomer	NK Ester 14G	40
Monomer	NK Oligo U-2PPA	20
Light stabilizer	Tinuvin 765	0.05
Thermal initiator	Trigonox 21S	1.0

The photochromic dyes were dissolved in NK Ester 2G first for three hours, then the remaining components were added in and stirred. After degassing and filtration, the monomer mix was filled in the mould assemblies and cured in an oven with the following heating profile:

Curing step	Curing program
Step 1	Start at 52 degree C
Step 2	Hold at 52 degree C for 3 hours
Step 3	Heat up to 53.5 degree C in 1.5 hours
Step 4	Heat up to 62.5 degree C in 4.5 hours
Step 5	Heat up to 80 degree C in 3.5 hours
Step 6	Hold at 80 degree C for 1 hours
Step 7	Heat up to 120 degree C in 5 hours
Step 8	Hold at 120 degree C for 2 hours
Step 9	Cool down to 80 degree C in 1 hour

This comparative example did not use terpinolene in the formulation. The cured lenses had optical distortions and were not uniform under polariscope and shadowscope. They were not considered to be useable as lenses.

Comparative example 4

A polymerisable composition containing the following components was prepared as described below:

Component Type	Component Name	Parts
Photochromic dye	Reversacol Pacific blue	0.08
Photochromic dye	Reversacol sun flower	0.017
Monomer	NK Ester 2G	36
Monomer	Light Ester 14 EG	31
Monomer	NK Oligo U-2PPA	33
Light stabilizer	Tinuvin 765	0.05
Thermal initiator	Trigonox 21S	0.4

The photochromic dyes were dissolved in NK Ester 2G first for three hours, then the remaining components were added in and stirred. After degassing and filtration, the monomer mix was filled in the moulds assemblies and cured in an oven with the following heating profile:

Curing step	Curing program
Step 1	Start at 52 degree C
Step 2	Hold at 52 degree C for 3 hours
Step 3	Heat up to 53.5 degree C in 1.5 hours
Step 4	Heat up to 62.5 degree C in 4.5 hours
Step 5	Heat up to 80 degree C in 4 hours
Step 6	Heat up to 90 degree C in 3.5 hours
Step 7	Hold at 90 degree C for 2 hours
Step 8	Cool down to 80 degree C in 1 hour
Step 9	Hold at 80 degree C for 1.5 hours

This comparative example did not use terpinolene in the formulation. The lenses obtained were not uniform under polariscope and shadowscope. The photochromic speed of the lenses was also much slower than the equivalent lenses produced using terpinolene.

Comparative example 5

A polymerisable composition containing the following components was prepared as described below:

Component Type	Component Name	Parts
Photochromic dye	Reversacol Pacific blue	0.08
Photochromic dye	Reversacol sunflower	0.017
Monomer	NK ester 2G	50
Monomer	Light ester 14 EG	30
Monomer	NK Oligo U2 PPA	20
Light stabilizer	Tinuvin 765	0.05
Thermal initiator	Trigonox 21S	0.4

The photochromic dyes were dissolved in NK Ester 2G first for three hours, then the remaining components were added in and stirred. After degassing and filtration, the monomer mix was filled in the moulds assemblies and cured in an oven with the following heating profile:

Curing step	Curing program
Step 1	Start at 52 degree C
Step 2	Hold at 52 degree C for 3 hours
Step 3	Heat up to 53.5 degree C in 1.5 hours
Step 4	Heat up to 62.5 degree C in 4.5 hours
Step 5	Hold at 90 degree C for 2 hours
Step 6	Cool down to 80 degree C in 1 hour
Step 7	Hold at 80 degree C for 1.5 hours

This comparative example did not use terpinolene in the formulation. The cured lenses were not uniform under polariscope and shadowscope and there were also a lot of strains in the lenses. These were not considered to be good lenses.

Comparative Example 6

A polymerisable composition containing the following components was prepared as described below:

Component Type	Component Name	Parts
Photochromic dye	CNN16	0.013
Photochromic dye	CNN12	0.0075
Photochromic dye	CNN13	0.04
Photochromic dye	CNN15	0.0075
Photochromic dye	CNN14	0.0075
Light Stabiliser	Tinuvin 765	0.05
Permanent dye	red dye concentrate(0.025% Solvaperm red BB in NK ester 2G)	0.04
Monomer	NK ester 2G	50

Monomer	NK ester 14G	30
Monomer	NK Oligo U-2PPA	20
Thermal initiator	Trigonox 21S	0.31

250 g of monomer mix was prepared based on the above formulation, the only difference between this example and example 6 is that there is no terpinolene in this example. After stirring, filtration and degassing, the monomer mix was filled into the following mould and gasket assemblies.

Moulds assemblies
Moulds for 15 mm thick semifinished lens
Moulds for 10mm thick semifinished lens
Moulds for +4.00 stock lens
Moulds for -2.00 stock lens
Moulds for 2 mm plano

The filled assemblies were placed in an oven and cured with the following curing cycle:

Curing step	Curing program
Step 1	Start at 52 °C
Step 2	Hold at 52 °C for 3 hours
Step 3	Heat up from 52 to 53.5 °C in 1.5 hours
Step 4	Heat up from 53.5 to 62.5 °C in 4.5 hours
Step 5	Heat up from 62.5 to 80 °C in 4 hours
Step 6	Hold at 80 °C for 8 hours

After curing the lenses were opened. The resulting lenses were of low quality and would not be acceptable for market. The following table summarises the quality issues of these lenses:

Lenses	Lens quality
15 mm thick semifinished lens	Massive surface defects due to the moulds separation during the curing step in the oven
10mm thick semifinished lens	Bubbles on the surface, massive surface defects.
+4.00 stock lens	Bubbles in the lens, massive surface defects due to the mould separation during the curing step in the oven
-2.00 stock lens	Surface defects
2 mm plano	Surface defects

The photochromic properties of the lenses were as follows:

Photochromic properties	
Initial T%	88.5%
Fully activated T%	14.4%
Photochromic speed, $T_{3/4}$ fade (seconds)	420 seconds

The resulting lenses had a Barcol hardness of 85.4.

Not only was the lens quality poor, the photochromic properties of the lenses were also poor. The fully activated darkness was low and the photochromic fading speed was slow.

Comparative Example 7

A polymerisable composition containing the following components was prepared as described below:

Component Type	Component Name	Parts
Photochromic dye	CNN16	0.013
Photochromic dye	CNN12	0.0075
Photochromic dye	CNN13	0.04
Photochromic dye	CNN15	0.0075
Photochromic dye	CNN14	0.0075
Light Stabiliser	Tinuvin 765	0.05
Permanent dye	red dye concentrate(0.025% Solvaperm red BB in NK ester 9G)	0.03
Monomer	NK ester 2G	30
Monomer	Light Ester 1.9ND	10
Monomer	NK ester 9G	30
Monomer	NK Oligo U-2PPA	30
Thermal initiator	Trigonox 21S	0.31

250 g of monomer mix was prepared based on the above formulation, the only difference between this example and Example 7 is that there is no terpinolene in this example. After stirring, filtration and degassing, the monomer mix is filled into following moulds and gasket assemblies:

Moulds assemblies
Moulds for 15 mm thick semifinished lens
Moulds for 10mm thick semifinished lens
Moulds for +4.00 stock lens
Moulds for -2.00 stock lens
Moulds for 2 mm plano

The filled assemblies were placed in the oven and cured with the following curing cycle:

Curing step	Curing program
Step 1	Start at 52 °C
Step 2	Hold at 52 °C for 3 hours
Step 3	Heat up from 52 to 53.5 °C in 1.5 hours
Step 4	Heat up from 53.5 to 62.5 °C in 4.5 hours
Step 5	Heat up from 62.5 to 80 °C in 4 hours
Step 6	Hold at 80 °C for 8 hours

After curing the lenses were opened. The resulting lenses were of low quality and would not be acceptable for market. The following table summarises the quality issues of these lenses:

Lenses	Lens quality
15 mm thick semifinished lens	Lens cracked into pieces
10mm thick semifinished lens	Massive surface defects.
+4.00 stock lens	Bubbles in the lens, massive surface defects due to the moulds separation during the curing step in the oven
-2.00 stock lens	Surface defects, lens cracked
2 mm plano	Surface waves

The photochromic properties of the lenses were as follows:

Initial T%	88.2%
Fully activated T%	15.4%
Photochromic speed, $T_{3/4}$ fade (seconds)	663 seconds

The resulting lenses had a Barcol hardness of 88.3.

As shown above, the photochromic speed of the lenses is very slow, and the lenses are poor in photochromic switching performance.

Comparative Example 8

A polymerisable composition containing the following components was prepared as described below:

Component Type	Component Name	Parts
Photochromic dye	CNN16	0.013
Photochromic dye	CNN12	0.005
Photochromic dye	CNN13	0.04
Photochromic dye	CNN15	0.0075
Photochromic dye	CNN14	0.01
Light Stabiliser	Tinuvin 765	0.05
Permanent dye	red dye concentrate(0.025% Solvaperm red BB in NK ester 9G)	0.03

Monomer	NK ester 2G	35
Monomer	NK ester 9G	20
Monomer	NK ester LA	10
Monomer	NK Oligo U-2PPA	35
Thermal initiator	Trigonox 21S	0.4

250 g of monomer mix was prepared based on the above formulation, the only difference between this example and Example 8 is that there is no terpinolene in this example. After stirring, filtration and degassing, the monomer mix was filled into following mould and gasket assemblies:

Moulds assemblies
Moulds for 15 mm thick semifinished lens
Moulds for 10mm thick semifinished lens
Moulds for +4.00 stock lens
Moulds for -2.00 stock lens
Moulds for 2 mm plano

The filled assemblies were placed in an oven and cured with the following curing cycle:

Curing step	Curing program
Step 1	Start at 52 °C
Step 2	Hold at 52 °C for 3 hours
Step 3	Heat up from 52 to 53.5 °C in 1.5 hours
Step 4	Heat up from 53.5 to 62.5 °C in 4.5 hours
Step 5	Heat up from 62.5 to 80 °C in 4 hours
Step 6	Hold at 80 °C for 8 hours

After curing the lenses were opened. The resulting lenses were of low quality and would not be acceptable for market. The following table summarises the quality issues of these lenses:

Lenses	Lens quality
15 mm thick semifinished lens	massive surface defects due to the moulds separation during the curing step in the oven
10mm thick semifinished lens	massive surface defects due to the moulds separation during the curing step in the oven
+4.00 stock lens	massive surface defects due to the moulds separation during the curing step in the oven
-2.00 stock lens	Surface defects
2 mm plano	Surface defects

The photochromic properties of the lenses were as follows:

Initial T%	88.4%
Fully activated T%	16.9%
Photochromic speed, $T_{3/4}$ fade (seconds)	888 seconds

The resulting lenses had a Barcol hardness of 86.4.

The lenses have poor photochromic properties as the photochromic switching speed is slow.

Comparative Example 9

A polymerisable composition containing the following components was prepared as described below:

Component Type	Component Name	Parts
Photochromic dye	CNN16	0.013
Photochromic dye	CNN12	0.0075
Photochromic dye	CNN13	0.04
Photochromic dye	CNN15	0.0075
Photochromic dye	CNN14	0.0075
Cure modifier	Terpinolene	0
Light Stabiliser	Tinuvin 765	0.05
Permanent dye	red dye concentrate(0.025% Solvaperm red BB in NK ester 2G)	0.04
Monomer	NK ester 2G	45
Monomer	NK ester 9PG	35
Monomer	NK Oligo U-2PPA	20
Thermal initiator	Trigonox 21S	0.31

250 g of monomer mix was prepared based on the above formulation, the only difference between this example and Example 9 is that there is no terpinolene in this example. After stirring, filtration and degassing, the monomer mix is filled into following mould and gasket assemblies:

Moulds assemblies	Number of assemblies
Moulds for 15 mm thick semifinished lens	1
Moulds for 10mm thick semifinished lens	1
Moulds for +4.00 stock lens	1
Moulds for -2.00 stock lens	1
Moulds for 2 mm plano	1

The filled assemblies were placed in an oven and cured with the following curing cycle:

Curing step	Curing program
Step 1	Start at 52 °C
Step 2	Hold at 52 °C for 3 hours
Step 3	Heat up from 52 to 53.5 °C in 1.5 hours
Step 4	Heat up from 53.5 to 62.5 °C in 4.5 hours
Step 5	Heat up from 62.5 to 80 °C in 4 hours
Step 6	Hold at 80 °C for 8 hours

After curing the lenses were opened. The resulting lenses were of low quality and would not be acceptable for market. The following table summarises the quality issues of these lenses:

Lenses	Lens quality
15 mm thick semifinished lens	cracked
10mm thick semifinished lens	cracked
+4.00 stock lens	Bubbles in the lens
-2.00 stock lens	Surface defects
2 mm plano	Surface defects

The photochromic properties of the lenses were as follows:

Photochromic properties	
Initial T%	88.0%
Fully activated T%	13.1%
Photochromic speed, $T_{3/4}$ fade (seconds)	557 seconds

The resulting lenses had a Barcol hardness of 88.1.

The lenses have poor photochromic properties as the photochromic fading speed is slow.

Comparative Example 10

A polymerisable composition containing the following components was prepared as described below:

Component Type	Component Name	Parts
Photochromic dye	CNN16	0.013
Photochromic dye	CNN12	0.0075
Photochromic dye	CNN13	0.04
Photochromic dye	CNN15	0.0075
Photochromic dye	CNN14	0.0075
Cure modifier	α -methyl styrene	0.5
Light Stabiliser	Tinuvin 765	0.05
Permanent dye	red dye concentrate(0.025% Solvaperm red BB in NK ester 2G)	0.04
Monomer	NK ester 2G	50
Monomer	NK ester 14G	30
Monomer	NK Oligo U-2PPA	20
Thermal initiator	Trigonox 21S	0.31

Instead of using Terpinolene as cure modifier, this example used α -methyl styrene in place of Terpinolene. All the other conditions were the same as Example 6.

250 g of monomer mix was prepared based on the above formulation. After stirring, filtration and degassing, the monomer mix was filled into following mould and gasket assemblies:

Moulds assemblies
Moulds for 15 mm thick semifinished lens
Moulds for 10mm thick semifinished lens
Moulds for +4.00 stock lens
Moulds for -2.00 stock lens
Moulds for 2 mm plano

The filled assemblies were placed in the oven and cured with the following curing cycle:

Curing step	Curing program
Step 1	Start at 52 °C
Step 2	Hold at 52 °C for 3 hours
Step 3	Heat up from 52 to 53.5 °C in 1.5 hours
Step 4	Heat up from 53.5 to 62.5 °C in 4.5 hours
Step 5	Heat up from 62.5 to 80 °C in 4 hours
Step 6	Hold at 80 °C for 8 hours

After curing the lenses were opened. The resulting lenses were of low quality and would not be acceptable for market. The following table summarises the quality issues of these lenses:

Lenses	Lens quality
15 mm thick semifinished lens	massive surface defects due to the moulds separation during the curing step in the oven
10mm thick semifinished lens	Cracked
+4.00 stock lens	cracked
-2.00 stock lens	Surface defects
2 mm plano	Surface defects

The photochromic properties of the lenses were as follows:

Initial T%	88.1%
Fully activated T%	14.4%
Photochromic speed, $T_{3/4}$ fade (seconds)	463 seconds

The resulting lenses had a Barcol hardness of 86.5.

The lenses have poor photochromic properties as the photochromic speed is slow.

Comparative Example 11

A polymerisable composition containing the following components was prepared as described below:

Component Type	Component Name	Parts
Photochromic dye	CNN16	0.013
Photochromic dye	CNN12	0.0075
Photochromic dye	CNN13	0.04
Photochromic dye	CNN15	0.0075
Photochromic dye	CNN14	0.0075
Cure modifier	α -methyl styrene	0.5
Light Stabiliser	Tinuvin 765	0.05
Permanent dye	red dye concentrate(0.025% Solvaperm red BB in NK ester 9G)	0.03
Monomer	NK ester 2G	30
Monomer	Light Ester 1.9ND	10
Monomer	NK ester 9G	30
Monomer	NK Oligo U-2PPA	30
Thermal initiator	Trigonox 21S	0.31

Instead of using Terpinolene as cure modifier, this example used α -methyl styrene in place of Terpinolene. All the other conditions were the same as Example 7.

250 g of monomer mix was prepared based on the above formulation. After stirring, filtration and degassing, the monomer mix is filled into following mould and gasket assemblies:

Moulds assemblies
Moulds for 15 mm thick semifinished lens
Moulds for 10mm thick semifinished lens
Moulds for +4.00 stock lens
Moulds for -2.00 stock lens
Moulds for 2 mm plano

The filled assemblies were placed in an oven and cured with the following curing cycle:

Curing step	Curing program
Step 1	Start at 52 °C
Step 2	Hold at 52 °C for 3 hours
Step 3	Heat up from 52 to 53.5 °C in 1.5 hours
Step 4	Heat up from 53.5 to 62.5 °C in 4.5 hours
Step 5	Heat up from 62.5 to 80 °C in 4 hours
Step 6	Hold at 80 °C for 8 hours

After curing the lenses were opened. The resulting lenses were of low quality and would not be acceptable for market. The following table summarises the quality issues of these lenses:

Lenses	Lens quality
15 mm thick semifinished lens	Massive cracks
10mm thick semifinished lens	Massive cracks
+4.00 stock lens	Massive cracks
-2.00 stock lens	massive surface defects due to the moulds separation during the curing step in the oven
2 mm plano	massive surface defects due to the moulds separation during the curing step in the oven

The photochromic properties of the lenses were as follows:

Initial T%	88.1%
Fully activated T%	17.1%
Photochromic speed, $T_{3/4}$ fade (seconds)	725 seconds

The resulting lenses had a Barcol hardness of 86.5.

The lenses have poor photochromic properties as the photochromic switching speed is very slow.

Comparative Example 12

A polymerisable composition containing the following components was prepared as described below:

Component Type	Component Name	Parts
Photochromic dye	CNN16	0.013
Photochromic dye	CNN12	0.005
Photochromic dye	CNN13	0.04
Photochromic dye	CNN15	0.0075
Photochromic dye	CNN14	0.01
Cure modifier	α -methyl styrene	0.5
Light Stabiliser	Tinuvin 765	0.05
Permanent dye	red dye concentrate(0.025% Solvaperm red BB in NK ester 9G)	0.03
Monomer	NK ester 2G	35
Monomer	NK ester 9G	20
Monomer	NK ester LA	10
Monomer	NK Oligo U-2PPA	35
Thermal initiator	Trigonox 21S	0.4

Instead of using Terpinolene as a cure modifier, this example used α -methyl styrene. All the other conditions were similar to Example 8.

250 g of monomer mix was prepared based on the above formulation. After stirring, filtration and degassing, the monomer mix was filled into following mould and gasket assemblies:

Moulds assemblies
Moulds for 15 mm thick semifinished lens
Moulds for 10mm thick semifinished lens
Moulds for +4.00 stock lens
Moulds for -2.00 stock lens
Moulds for 2 mm plano

The filled assemblies were placed in the oven and cured with the following curing cycle:

Curing step	Curing program
Step 1	Start at 52 °C
Step 2	Hold at 52 °C for 3 hours
Step 3	Heat up from 52 to 53.5 °C in 1.5 hours
Step 4	Heat up from 53.5 to 62.5 °C in 4.5 hours
Step 5	Heat up from 62.5 to 80 °C in 4 hours
Step 6	Hold at 80 °C for 8 hours

After curing the lenses were opened. The resulting lenses were of low quality and would not be acceptable for market. The following table summarises the quality issues of these lenses:

Lenses	Lens quality
15 mm thick semifinished lens	Massive cracks
10mm thick semifinished lens	Massive cracks
+4.00 stock lens	massive surface defects due to the moulds separation during the curing step in the oven
-2.00 stock lens	massive surface defects due to the moulds separation during the curing step in the oven
2 mm plano	massive surface defects due to the moulds separation during the curing step in the oven

The photochromic properties of the lenses were as follows:

Initial T%	86.8%
Fully activated T%	16.0%
Photochromic speed, $T_{3/4}$ fade (seconds)	833 seconds

The resulting lenses had a Barcol hardness of 87.3.

The lenses have poor photochromic properties as the photochromic switching speed is very slow.

Comparative Example 13

A polymerisable composition containing the following components was prepared as described below:

Component Type	Component Name	Parts
Photochromic dye	CNN16	0.013
Photochromic dye	CNN12	0.0075
Photochromic dye	CNN13	0.04
Photochromic dye	CNN15	0.0075
Photochromic dye	CNN14	0.0075
Cure modifier	α -methyl styrene	0.5
Light Stabiliser	Tinuvin 765	0.05
Permanent dye	red dye concentrate(0.025% Solvaperm red BB in NK ester 2G)	0.04
Monomer	NK ester 2G	45
Monomer	NK ester 9PG	35
Monomer	NK Oligo U-2PPA	20
Thermal initiator	Trigonox 21S	0.31

Instead of using Terpinolene as cure modifier, this example used α -methyl styrene. All the other conditions were similar to Example 9.

250 g of monomer mix was prepared based on the above formulation. After stirring, filtration and degassing, the monomer mix was filled into the following mould and gasket assemblies:

Moulds assemblies
Moulds for 15 mm thick semifinished lens
Moulds for 10mm thick semifinished lens
Moulds for +4.00 stock lens
Moulds for -2.00 stock lens
Moulds for 2 mm plano

The filled assemblies were placed in the oven and cured with the following curing cycle:

Curing step	Curing program
Step 1	Start at 52 °C
Step 2	Hold at 52 °C for 3 hours
Step 3	Heat up from 52 to 53.5 °C in 1.5 hours
Step 4	Heat up from 53.5 to 62.5 °C in 4.5 hours
Step 5	Heat up from 62.5 to 80 °C in 4 hours
Step 6	Hold at 80 °C for 8 hours

After curing the lenses were opened. The resulting lenses were of low quality and would not be acceptable for market. The following table summarises the quality issues of these lenses:

Lenses	Lens quality
15 mm thick semifinished lens	A 2 cm long crack, massive surface defects
10mm thick semifinished lens	massive surface defects due to the moulds separation during the curing step in the oven
+4.00 stock lens	massive surface defects due to the moulds separation during the curing step in the oven
-2.00 stock lens	massive surface defects due to the moulds separation during the curing step in the oven
2 mm plano	massive surface defects due to the moulds separation during the curing step in the oven

The photochromic properties of the lenses were as follows:

Initial T%	87.8%
Fully activated T%	13.4%
Photochromic speed, $T_{3/4}$ fade (seconds)	549 seconds

The resulting lenses had a Barcol hardness of 89.1.

The lenses have poor photochromic properties as the photochromic switching speed is slow.

Comparative Example 14

A polymerisable composition containing the following components was prepared as described below:

Component Type	Component Name	Parts
Photochromic dye	CNN16	0.013
Photochromic dye	CNN12	0.0075
Photochromic dye	CNN13	0.04
Photochromic dye	CNN15	0.0075
Photochromic dye	CNN14	0.0075
Cure modifier	1-dodecane thiol	0.5
Light Stabiliser	Tinuvin 765	0.05
Permanent dye	red dye concentrate(0.025% Solvaperm red BB in NK ester 2G)	0.04
Monomer	NK ester 2G	50
Monomer	NK ester 14G	30
Monomer	NK Oligo U-2PPA	20
Thermal initiator	Trigonox 21S	0.31

Instead of using Terpinolene as cure modifier, this example used 1-dodecane thiol. All the other conditions were similar to Example 6.

250 g of monomer mix was prepared based on the above formulation. After stirring, filtration and degassing, the monomer mix was filled into the following mould and gasket assemblies:

Moulds assemblies
Moulds for 15 mm thick semifinished lens
Moulds for 10mm thick semifinished lens
Moulds for +4.00 stock lens
Moulds for -2.00 stock lens
Moulds for 2 mm plano

The filled assemblies were placed in the oven and cured with the following curing cycle:

Curing step	Curing program
Step 1	Start at 52 °C
Step 2	Hold at 52 °C for 3 hours
Step 3	Heat up from 52 to 53.5 °C in 1.5 hours
Step 4	Heat up from 53.5 to 62.5 °C in 4.5 hours
Step 5	Heat up from 62.5 to 80 °C in 4 hours
Step 6	Hold at 80 °C for 8 hours

After curing the lenses were opened. The resulting lenses were of low quality and would not be acceptable for market. The following table summarises the quality issues of these lenses:

Lenses	Lens quality
15 mm thick semifinished lens	A 3 cm long crack, massive surface defects
10mm thick semifinished lens	A 2 cm long crack, massive surface defects
+4.00 stock lens	A 5 cm long crack, massive surface defects
-2.00 stock lens	surface defects due to the moulds separation during the curing step in the oven
2 mm plano	surface defects due to the moulds separation during the curing step in the oven

The photochromic properties of the lenses were as follows:

Initial T%	87.7%
Fully activated T%	14.7%
Photochromic speed, $T_{3/4}$ fade (seconds)	446 seconds

The resulting lenses had a Barcol hardness of 87.2.

The lenses have poor photochromic properties as the photochromic switching speed is slow.

Comparative Example 15

A polymerisable composition containing the following components was prepared as described below:

Component Type	Component Name	Parts
Photochromic dye	CNN16	0.013
Photochromic dye	CNN12	0.0075
Photochromic dye	CNN13	0.04
Photochromic dye	CNN15	0.0075
Photochromic dye	CNN14	0.0075
Cure modifier	1-dodecane thiol	0.5
Light Stabiliser	Tinuvin 765	0.05
Permanent dye	red dye concentrate(0.025% Solvaperm red BB in NK ester 9G)	0.03
Monomer	NK ester 2G	30
Monomer	Light Ester 1.9ND	10
Monomer	NK ester 9G	30
Monomer	NK Oligo U-2PPA	30
Thermal initiator	Trigonox 21S	0.31

Instead of using Terpinolene as cure modifier, this example used 1-dodecane thiol. All the other conditions were similar to Example 7.

250 g of monomer mix was prepared based on the above formulation. After stirring, filtration and degassing, the monomer mix was filled into following mould and gasket assemblies:

Moulds assemblies
Moulds for 15 mm thick semifinished lens
Moulds for 10mm thick semifinished lens
Moulds for +4.00 stock lens
Moulds for -2.00 stock lens
Moulds for 2 mm plano

The filled assemblies were placed in the oven and cured with the following curing cycle:

Curing step	Curing program
Step 1	Start at 52 °C
Step 2	Hold at 52 °C for 3 hours
Step 3	Heat up from 52 to 53.5 °C in 1.5 hours
Step 4	Heat up from 53.5 to 62.5 °C in 4.5 hours
Step 5	Heat up from 62.5 to 80 °C in 4 hours
Step 6	Hold at 80 °C for 8 hours

After curing the lenses were opened. The resulting lenses were of low quality and would not be acceptable for market. The following table summarises the quality issues of these lenses:

Lenses	Lens quality
15 mm thick semifinished lens	A 1 cm long crack, massive surface defects
10mm thick semifinished lens	three crack s in the lens, massive surface defects
+4.00 stock lens	A 1cm long crack, massive surface defects
-2.00 stock lens	Surface defects
2 mm plano	surface defects

The photochromic properties of the lenses were as follows:

Initial T%	88.3%
Fully activated T%	17.0%
Photochromic speed, $T_{3/4}$ fade (seconds)	724 seconds

The resulting lenses had a Barcol hardness of 88.2.

The lenses have poor photochromic properties as the photochromic switching speed is very slow.

Comparative Example 16

A polymerisable composition containing the following components was prepared as described below:

Component Type	Component Name	Parts
Photochromic dye	CNN16	0.013
Photochromic dye	CNN12	0.005
Photochromic dye	CNN13	0.04
Photochromic dye	CNN15	0.0075
Photochromic dye	CNN14	0.01
Cure modifier	1-dodecane thiol	0.5
Light Stabiliser	Tinuvin 765	0.05
Permanent dye	red dye concentrate(0.025% Solvaperm red BB in NK ester 9G)	0.03
Monomer	NK ester 2G	35
Monomer	NK ester 9G	20
Monomer	NK ester LA	10
Monomer	NK Oligo U-2PPA	35
Thermal initiator	Trigonox 21S	0.4

Instead of using Terpinolene as cure modifier, this example used 1-dodecane thiol. All the other conditions were similar to Example 8.

250 g of monomer mix was prepared based on the above formulation. After stirring, filtration and degassing, the monomer mix was filled into the following mould and gasket assemblies:

Mould assemblies
Moulds for 15 mm thick semifinished lens
Moulds for 10mm thick semifinished lens
Moulds for +4.00 stock lens
Moulds for -2.00 stock lens
Moulds for 2 mm plano

The filled assemblies were placed in the oven and cured with the following curing cycle:

Curing step	Curing program
Step 1	Start at 52 °C
Step 2	Hold at 52 °C for 3 hours
Step 3	Heat up from 52 to 53.5 °C in 1.5 hours
Step 4	Heat up from 53.5 to 62.5 °C in 4.5 hours
Step 5	Heat up from 62.5 to 80 °C in 4 hours
Step 6	Hold at 80 °C for 8 hours

After curing the lenses were opened. The resulting lenses were of low quality and would not be acceptable for market. The following table summarises the quality issues of these lenses:

Lenses	Lens quality
15 mm thick semifinished lens	A 1 cm long crack, massive surface defects
10mm thick semifinished lens	three cracks in the lens, massive surface defects
+4.00 stock lens	A 1cm long crack, massive surface defects
-2.00 stock lens	Surface defects
2 mm plano	surface defects

The photochromic properties of the lenses were as follows:

Photochromic properties	
Initial T%	87.7%
Fully activated T%	17.4%
Photochromic speed, $T_{3/4}$ fade (seconds)	956 seconds

The resulting lenses had a Barcol hardness of 87.1.

The lenses have poor photochromic properties as the photochromic switching speed is slow.

Comparative Example 17

A polymerisable composition containing the following components was prepared as described below:

Component Type	Component Name	Parts
Photochromic dye	CNN16	0.013
Photochromic dye	CNN12	0.0075
Photochromic dye	CNN13	0.04
Photochromic dye	CNN15	0.0075
Photochromic dye	CNN14	0.0075
Cure modifier	1-dodecane thiol	0.5
Light Stabiliser	Tinuvin 765	0.05
Permanent dye	red dye concentrate(0.025% Solvaperm red BB in NK ester 2G)	0.04
Monomer	NK ester 2G	45
Monomer	NK ester 9PG	35
Monomer	NK Oligo U-2PPA	20
Thermal initiator	Trigonox 21S	0.31

Instead of using Terpinolene as cure modifier, this example used 1-dodecane thiol. All the other conditions were similar to Example 9.

250 g of monomer mix was prepared based on the above formulation. After stirring, filtration and degassing, the monomer mix was filled into the following mould and gasket assemblies:

Mould assemblies
Moulds for 15 mm thick semifinished lens
Moulds for 10mm thick semifinished lens
Moulds for +4.00 stock lens
Moulds for -2.00 stock lens
Moulds for 2 mm plano

The filled assemblies were placed in an oven and cured with the following curing cycle:

Step Number	Step Description
Step 1	Start at 52 °C
Step 2	Hold at 52 °C for 3 hours
Step 3	Heat up from 52 to 53.5 °C in 1.5 hours
Step 4	Heat up from 53.5 to 62.5 °C in 4.5 hours
Step 5	Heat up from 62.5 to 80 °C in 4 hours
Step 6	Hold at 80 °C for 8 hours

After curing the lenses were opened. The resulting lenses were of low quality and would not be acceptable for market. The following table summarises the quality issues of these lenses:

Lenses	Lens quality
15 mm thick semifinished lens	massive cracks, massive surface defects
10mm thick semifinished lens	massive surface defects
+4.00 stock lens	Massive surface ring marks
-2.00 stock lens	One separation mark on the lens front surface
2 mm plano	Lens was okay

The photochromic properties of the lenses were as follows:

Initial T%	87.5%
Fully activated T%	15.5%
Photochromic speed, $T_{3/4}$ fade (seconds)	670 seconds

The resulting lenses had a Barcol hardness of 89.5.

The lenses have poor photochromic properties as the photochromic switching speed was very slow.

Comparative Example 18

A polymerisable composition containing the following components was prepared as described below:

Component Type	Component Name	Parts
Cure modifier	Terpinolene	0.5
Light Stabiliser	Tinuvin 765	0.05
Permanent dye	red dye concentrate(0.025% Solvaperm red BB in NK ester 2G)	0.04
Monomer	NK ester 2G	50
Monomer	NK ester 14G	30
Monomer	NK Oligo U-2PPA	20
Thermal initiator	Trigonox 21S	0.31

Monomer mix was prepared based on above formulation. The monomer mix was filled into mould assemblies and curing in an oven with the following curing profile:

Curing step	Curing program
Step 1	Start at 52 °C
Step 2	Hold at 52 °C for 3 hours
Step 3	Heat up from 52 to 53.5 °C in 1.5 hours
Step 4	Heat up from 53.5 to 62.5 °C in 4.5 hours

Step 5	Heat up from 62.5 to 80 °C in 4 hours
Step 6	Hold at 80 °C for 8 hours

After curing the lenses were opened. The resulting lenses had a Barcol hardness of 76.5 and no photochromic activity. In order to achieve photochromic activity, another expensive process step has to be introduced to add the photochromic activity into these lenses. These extra steps could be the application of photochromic coating or performance of photochromic imbibition methods. However, these are undesirable as they significantly increase the cost of the process.

Summary of Exemplary Examples

The following table provides a summary of exemplary examples and exemplary comparative examples discussed above:

Example 6 0.5% terpinolene <i>Good lenses</i>	Comparative Example 6 No terpinolene <i>Poor lenses</i>	Comparative Example 10 0.5% α -methyl styrene <i>Poor lenses</i>	Comparative Example 14 0.5% 1-dodecane thiol <i>Poor lenses</i>
Example 7 0.5% terpinolene <i>Good lenses</i>	Comparative Example 7 No terpinolene <i>Poor lenses</i>	Comparative Example 11 0.5% α -methyl styrene <i>Poor lenses</i>	Comparative Example 15 0.5% 1-dodecane thiol <i>Poor lenses</i>
Example 8 0.5% terpinolene <i>Good lenses</i>	Comparative Example 8 No terpinolene <i>Poor lenses</i>	Comparative Example 12 0.5% α -methyl styrene <i>Poor lenses</i>	Comparative Example 16 0.5% 1-dodecane thiol <i>Poor lenses</i>
Example 9 0.5% terpinolene <i>Good lenses</i>	Comparative Example 9 No terpinolene <i>Poor lenses</i>	Comparative Example 13 0.5% α -methyl styrene <i>Poor lenses</i>	Comparative Example 17 0.5% 1-dodecane thiol <i>Poor lenses</i>

As shown above, the use of terpinolene as a cure modifier in polymerisable compositions results in lenses of a higher quality than those without a cure modifier or those that use α -methyl styrene or 1-dodecane thiol as a cure modifier.

Those skilled in the art will appreciate that the invention described herein is susceptible to variations and modifications other than those specifically described. It is to be understood that the invention includes all such variations and modifications. The invention also includes all of the steps, features, compositions and compounds referred to, or indicated in this specification, individually or collectively, and any and all combinations of any two or more of the steps or features.

The claims defining the invention are as follows:

1. A liquid casting composition for producing photochromic optical elements, the composition including:
 - (i) an acrylate or methacrylate monomer,
 - (ii) a photochromic compound,
 - (iii) a terpinolene-based radical chain transfer agent, and
 - (iv) a thermal free radical initiator.

2. A composition according to claim 1, wherein the acrylate or methacrylate monomer includes one or more monomers selected from the group consisting of: alkylene acrylates; alkylene methacrylates; poly(alkyleneoxide) acrylates; poly(alkyleneoxide) methacrylates; urethane acrylates; and urethane methacrylates.

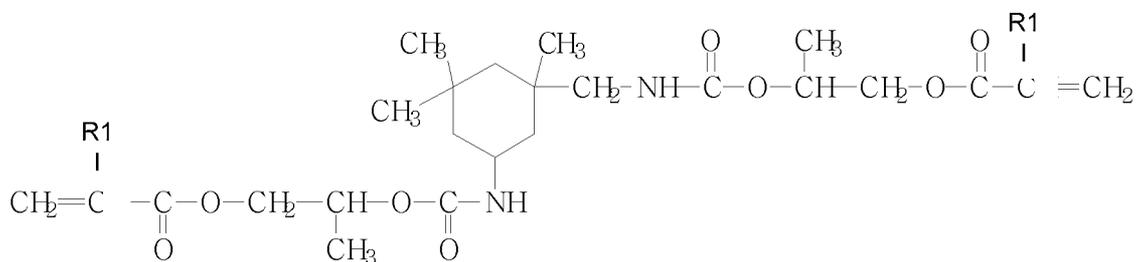
3. A composition according to claim 2, wherein the one or more monomers include one or more monoacrylates, monomethacrylates, diacrylates, dimethacrylates, triacrylates or trimethacrylates.

4. A composition according to any one of claims 1 to 3, wherein the acrylate or methacrylate monomer includes one or more monomers selected from the group consisting of: decanediol diacrylate; NK Ester A200 (polyethylene glycol 200 diacrylate); NK Ester A400 (polyethylene glycol 400 diacrylate); NK Ester A600 (polyethylene glycol 600 diacrylate); NK Ester APG-200 (tripropylene glycol diacrylate); NK Ester APG-400 (polypropylene glycol 400 diacrylate); NK Ester 14G; NK Ester 9G; NK Ester 9PG; NK Ester 4G; NK Ester 2G; NK Oligo U-4HA; NK Oligo U-6HA; NK Oligo U-2PPA and NK Ester LA.

5. A composition according to any one of claims 1 to 4, wherein the acrylate or methacrylate monomer includes one or more monomers selected from the group consisting of: NK Ester 14G; NK Ester 9PG; NK Ester 9G; NK Ester 2G; 1,12-Dodecanediol Dimethacrylate, 1.10-Decanediol Di Methacrylate; 1.9-Nonanediol Di Methacrylate (Light Ester 1.9 ND); Lauryl (meth)acrylate (NK Ester LA); Isostearyl (meth)acrylate; and an ethoxylated bisphenol-A dimethacrylate having between 1 and 10 ethoxy groups per molecule.

6. A composition according to any one of claims 1 to 5, wherein the acrylate or methacrylate monomer also contains a urethane acrylate or a urethane methacrylate monomer.

7. A composition according to claim 6, wherein the urethane acrylate or a urethane methacrylate monomer includes one or more monomers selected from the group consisting of: NK Oligo U-4HA, NK Oligo U-6HA, and a monomer of the following structure:



wherein R1 represents a hydrogen atom or a methyl group.

8. A composition according to any one of claims 1 to 7, wherein the acrylate or methacrylate monomer includes a combination of:

- a relatively hard polymer formed from a polyalkylene oxide acrylate or methacrylate monomer;
- a relatively flexible polymer formed from a polyalkylene oxide acrylate or methacrylate monomer; and
- a urethane acrylate or methacrylate monomer;

9. A composition according to claim 8, wherein the relatively hard polymer is formed from diethyleneglycol dimethacrylate or a similar monomer.

10. A composition according to claim 8 or claim 9, wherein the relatively flexible polymer is formed from a monomer having 14 alkyleneoxide units per monomer molecule.

11. A composition according to any one of claims 8 to 10, wherein the relatively flexible polymer is formed from Light Ester 14 EG or a similar monomer.

12. A composition according to any one of claims 1 to 7, wherein the acrylate or methacrylate monomer includes a combination of:

- a relatively flexible polymer formed from a polyalkylene oxide acrylate or methacrylate monomer; a polypropylene glycol acrylate or methacrylate monomer; a lauryl acrylate or methacrylate monomer; or a isostearyl acrylate or methacrylate monomer;
- a relatively hard polymer formed from a bisphenol A ethoxylated diacrylate or methacrylate; and
- a urethane acrylate or methacrylate monomer.

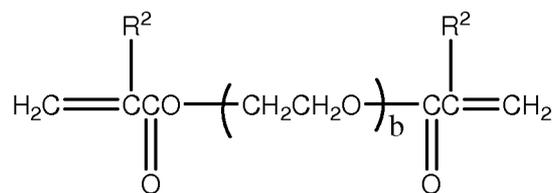
13. A composition according to claim 12, wherein the relatively flexible polymer is formed from a monomer having 9 alkyleneoxide units per monomer molecule.

14. A composition according to claim 12 or claim 13, wherein the relatively flexible polymer is formed from NK Ester 9G or a similar monomer.

15. A composition according to claim 12 or claim 13, wherein the relatively flexible polymer is formed from NK Ester 9PG or a similar monomer.

16. A composition according to any one of claims 1 to 15, wherein the composition includes:

- 30 to 60 parts by weight of a polymerisable composition polyethylene glycol di(meth)acrylate monomer of formula (II):



(II)

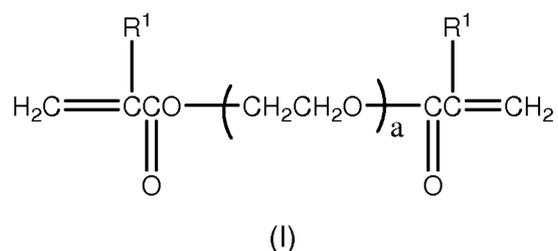
wherein R² is a hydrogen atom or a methyl group, and b is an integer selected from the group consisting of 5 to 20,

- 30 to 60 parts by weight of a bisphenol A ethoxylated di(meth)acrylate;
- 0 to 40 parts by weight of the polymerisable composition of a urethane (meth)acrylate monomer;
- 0.02 to 0.2 parts by weight of the polymerisable composition of the photochromic compound;

- 0.1 to 1.0 parts by weight of the polymerisable composition of the terpinolene-based radical chain transfer agent; and
- 0.1 to 1.5 parts by weight of the polymerisable composition of the thermal free radical initiator.

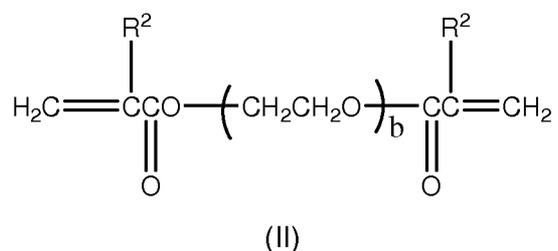
17. A composition according to any one of claims 1 to 15, wherein the composition includes:

- 20 to 60 parts by weight of the polymerisable composition of a polyethylene glycol di(meth)acrylate monomer of formula (I):



wherein R¹ is a hydrogen atom or a methyl group, and a is an integer selected from the group consisting of 2 to 4;

- 15 to 60 parts by weight of the polymerisable composition of a polyethylene glycol di(meth)acrylate monomer of formula (II):



wherein R² is a hydrogen atom or a methyl group, and b is an integer selected from the group consisting of 5 to 20;

- 0 to 40 parts by weight of the polymerisable composition of a urethane (meth)acrylate monomer;
- 0.02 to 0.2 parts by weight of the polymerisable composition of the photochromic compound;
- 0.1 to 1.0 parts by weight of the polymerisable composition of the terpinolene-based radical chain transfer agent; and

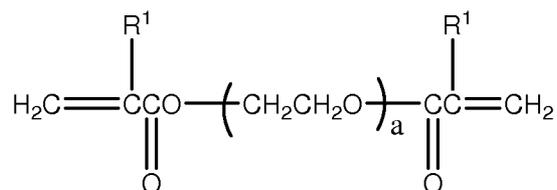
- 0.1 to 1.5 parts by weight of the polymerisable composition of the thermal free radical initiator.

18. A composition according to claim 17, wherein the composition further comprises 5 to 20% parts by weight of the polymerisable composition of 1.9 nonanediol dimethacrylate.

19. A composition according to claim 17, wherein the composition further comprises 5 to 20% parts by weight of the polymerisable composition of n-Lauryl methacrylate.

20. A composition according to any one of claims 1 to 15, wherein the composition includes:

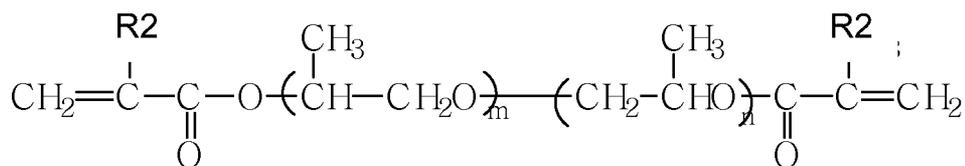
- 30 to 50 parts by weight of the polymerisable composition a polyethylene glycol di(meth)acrylate monomer of formula (I):



(I)

wherein R¹ is a hydrogen atom or a methyl group, and a is an integer selected from the group consisting of 2 to 4;

- 30 to 60 parts by weight of the polymerisable composition of a polypropylene glycol di(meth)acrylate monomer of formula (II):



(II)

wherein R2 represents a hydrogen atom or a methyl group, and m and n are integers that together add up to 7;

- 0 to 40 parts by weight of the polymerisable composition of a urethane (meth)acrylate monomer;

- 0.02 to 0.2 parts by weight of the polymerisable composition of the photochromic compound;
- 0.1 to 1.0 parts by weight of the polymerisable composition of the terpinolene-based radical chain transfer agent; and
- 0.1 to 1.5 parts by weight of the polymerisable composition of the thermal free radical initiator.

21. A composition according to any one of claims 1 to 20, wherein the photochromic compound includes one or more photochromic compounds selected from the group consisting of: spiro-naphthoxazines; spiropyrans, naphthopyrans; triarylmethanes, stilbenes, azastilbenes, nitrones, fulgides, spiropyrans, spiro-oxazines, quinones; and chromenes.

22. A composition according to any one of claims 1 to 21, wherein the terpinolene-based radical chain transfer agent includes one or more chain transfer agents selected from the group consisting of: terpinolene, 1-isopropyl-4-methyl-1,4-cyclohexadiene, and 1-isopropyl-4-methyl-1,3-cyclohexadiene.

23. A composition according to any one of claims 1 to 22, wherein the terpinolene-based radical chain transfer agent is added to the polymerisable composition in an amount from 0.01 percent to 2 percent by weight based on the total weight of the polymerisable composition.

24. A composition according to any one of claims 1 to 23, wherein the terpinolene-based radical chain transfer agent is added to the polymerisable composition in an amount from 0.01 percent to 1 percent by weight based on the total weight of the polymerisable composition.

25. A composition according to any one of claims 1 to 24, wherein the thermal free radical initiator includes an organic peroxide thermal free radical initiator or an azo free radical initiator.

26. A composition according to any one of claims 1 to 24, wherein the thermal free radical initiator includes tert-Butyl peroxy-2-ethylhexanoate.

27. A composition according to any one of claims 1 to 26, wherein the composition contains one or more additional adjuvants.
28. A composition according to claim 27, wherein the one or more additional adjuvants include a light stabiliser and/or UV absorber.
29. A composition according to claim 28, wherein the light stabiliser includes a hindered amine light stabilizer.
30. A composition according to claim 28 or claim 29, wherein the UV absorber includes a member or derivate of a benzophenone class, benzotriazole class, cyanoacrylate class, triazine class, or a combination of any of the foregoing.
31. A composition according to any one of claims 27 to 30, wherein the one or more additional adjuvants include an antioxidant.
32. A composition according to claim 31, wherein the antioxidant includes a butylated hydroxytoluene (BHT), a hindered phenol, a phosphate, a thioester or a combination of any of the foregoing.
33. A composition according to any one of claims 27 to 32, wherein the one or more additional adjuvants include a permanent dye.
34. A composition according to claim 33, wherein the permanent dye is included in a quantity to mask residual colour in the composition and/or optical element without otherwise colouring the composition and/or optical element.
35. A composition according to any one of claims 1 to 34, wherein the composition is suitable for forming photochromic optical elements with a Barcol hardness of between 60 and 80.
36. A composition according to any one of claims 1 to 35, wherein the composition is suitable for forming photochromic optical elements with a photochromic $T_{3/4}$ fade time less than 300 seconds.

37. A composition according to any one of claims 1 to 36, wherein the composition is suitable for forming photochromic optical elements with a photochromic $T_{3/4}$ fade time less than 250 seconds.

38. A composition according to any one of claims 1 to 37, wherein the composition is suitable for forming first and second crack-free and surface defect-free photochromic optical elements with centre thickness between 1.5 mm and 17 mm, wherein the centre thickness of the first optical element is at least 6 mm greater than that of the second optical element.

39. A composition according to any one of claims 1 to 38, wherein the composition is suitable for forming first and second crack-free and surface defect-free photochromic optical elements with centre thickness between 1.5 mm and 17 mm, wherein the centre thickness of the first optical element is at least 12 mm greater than that of the second optical element.

40. A process for the manufacture of a photochromic optical element, the process including:

- introducing the composition according to any one of claims 1 to 38 into a mould; and
- subjecting the composition to a thermal curing step to provide a solid photochromic optical element.

41. A process according to claim 40, wherein the thermal curing step includes raising the temperature of the composition from around 50°C to between 80°C and 120°C over a period of 7 to 14 hours.

42. A process according to claim 40 or claim 41, wherein the thermal curing step includes:

- heating the composition at 52°C for 3 hours;
- heating the composition from 52°C to 53.5°C over 1.5 hours;
- heating the composition from 53.5°C to 62.5°C over 4.5 hours;
- heating the composition from 62.5°C to 80°C over 4 hours; and
- maintaining the temperature at 80°C for 8 hours;

43. An optical element formed from a composition according to any one of claims 1 to 39 or according to a process of any one of claims 40 to 42.
44. An optical element according to claim 43, wherein the optical element is a lens.
45. An optical element according to claim 42, wherein the lens is an ophthalmic lens.
46. An optical element according to any one of claims 43 to 45, wherein the optical element has a photochromic $T_{3/4}$ fade time less than 300 seconds.
47. An optical element according to any one of claims 43 to 46, wherein the optical element has a photochromic $T_{3/4}$ fade time less than 250 seconds.
48. An optical element according to any one of claims 43 to 47, wherein the optical element has a Barcol hardness of between 60 and 80.
49. An optical element according to any one of claims 43 to 48, wherein the optical element has a centre thickness of between 1.5 mm and 17 mm.
50. A liquid casting composition as described herein and substantially as hereinbefore described with respect to the accompanying examples.
51. A process as described herein and substantially as hereinbefore described with respect to the accompanying examples.
52. An optical element as described herein and substantially as hereinbefore described with respect to the accompanying examples.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/AU2010/000297

A. CLASSIFICATION OF SUBJECT MATTER		
Int. Cl.		
<i>C08F 2/44</i> (2006.01)	<i>C08F 20/26</i> (2006.01)	
<i>B29D 11/00</i> (2006.01)	<i>G02B 1/00</i> (2006.01)	
According to International Patent Classification (IPC) or to both national classification and IPC		
B. FIELDS SEARCHED		
Minimum documentation searched (classification system followed by classification symbols)		
Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched		
Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)		
EPODOC; WPI; CAPLUS & GOOGLE: photo_chromi+, +acrylate+, terpinolene+, lens, optical, ophthal+		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 2008/153595 A1 (SOFTLIGHT, INC.) 18 December 2008 (see pg. 6, 2 nd last para.; pg. 8, 3 rd -4 th para.; pg. 9, 2 nd para.; pg. 14, last para.; Examples, claims)	1-18, 20-34, 38-41, 43-45 19, 49
Y		
X	US 2003/0043447 A1 (TOH et al.) 6 March 2003 (see Abstract, para. 0001, 0002, 0017, 0018, 0024, 0027, 0028, 0034, 0039, 0077, Tables 1-6; Examples)	1-16, 18, 20-29, 33-40, 43-48 19, 49
Y		
X	US 2005/0116381 A1 (WONG et al.) 2 June 2005 (see Abstract, para. 0038, 0039, 0050-0081, Table 2, Examples, claims)	1-16, 18, 20-27, 33, 29, 33, 35- 41, 43-48 19, 49
Y		
<input checked="" type="checkbox"/> Further documents are listed in the continuation of Box C		<input checked="" type="checkbox"/> See patent family annex
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"P" document published prior to the international filing date but later than the priority date claimed		
Date of the actual completion of the international search 11 May 2010	Date of mailing of the international search report 14 MAY 2010	
Name and mailing address of the ISA/AU AUSTRALIAN PATENT OFFICE PO BOX 200, WODEN ACT 2606, AUSTRALIA E-mail address: pct@ipaaustralia.gov.au Facsimile No. +61 2 6283 7999	Authorized officer SALESH SWAMI AUSTRALIAN PATENT OFFICE (ISO 9001 Quality Certified Service) Telephone No : +61 2 6283 7939	

INTERNATIONAL SEARCH REPORT

International application No.

PCT/AU2010/000297

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
L	US 5373033 A (TOH et al.) 13 December 1994 (see Example 6)	1-34, 38-41, 43-45, 49
Y	US 3565814 A (PELLON) 23 February 1971 (see Example 1)	19
Y	US 6365312 B1 (FOUCHER et al.) 2 April 2002 (see Examples)	19
Y	US 2005/0258408 A1 (MOLOCK et al.) 24 November 2005 (see claims)	19
Y	US 2008/0311287 A1 (CHEN) 18 December 2008 (see para. 006)	49
Y	US 5696621 A (NAKANISHI et al.) 9 December 1997 (see Abstract, col. 7, lines 47-50)	49

Box No. II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

2. Claims Nos.: **50-52**
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:

Claims 50-52 do not comply with Rule 6.2(a) because they rely on references to the description and/or drawings

3. Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a)

Box No. III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. As all searchable claims could be searched without effort justifying additional fees, this Authority did not invite payment of additional fees.
3. As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:

4. No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

- The additional search fees were accompanied by the applicant's protest and, where applicable, the payment of a protest fee.
- The additional search fees were accompanied by the applicant's protest but the applicable protest fee was not paid within the time limit specified in the invitation.
- No protest accompanied the payment of additional search fees.

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.

PCT/AU2010/000297

This Annex lists the known "A" publication level patent family members relating to the patent documents cited in the above-mentioned international search report. The Australian Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

Patent Document Cited in Search Report		Patent Family Member					
WO	2008153595	NONE					
US	2003043447	AU	43955/01	BR	0109657	CA	2403391
		EP	1276774	MX	PA02009496	US	6811830
		WO	0172851				
US	2005116381	AU	2002366250	BR	0214893	CN	1604841
		EP	1453662	MX	PA04005777	WO	03051615
US	5373033	AU	75160/91	EP	0453149	HK	103195
		JP	4227617				
US	3565814	NONE					
US	6365312	EP	1260872	JP	2003057785	US	6458165
US	2005258408	AR	032841	AR	055572	AU	15676/02
		AU	2006234938	BR	0200519	BR	PI0607036
		CA	2371965	CA	2603510	CN	1374356
		CN	101189536	EP	1235096	EP	1869506
		HK	1046554	HK	1049181	JP	2002333600
		KR	20020069156	KR	20080011661	SG	155758
		US	2002133889	US	2003000028	WO	2006110306
US	2008311287	AU	2006326863	EP	1963889	WO	2007070975
US	5696621	JP	7287181				
Due to data integration issues this family listing may not include 10 digit Australian applications filed since May 2001.							
END OF ANNEX							